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04 MAR 2008

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Milbank, Tweed, Hadley & McCloy
1850 K St., NW Site 1100
Washington, DC 20009

Dear Mr. Longton:

On 11 July 2007, you filed in the United States Patent and Trademark Office (USPTO) papers requesting entry into the national phase of international application number PCT/EP2005/000221 filed 12 January 2005, entitled Extrusion Coating Polyethylene. The inventors are indicated as Laiho, Erkki; Sainio, Markku; and Haapaniemi, Karoliina. Your attorney docket number is indicated as 37488-01300.

Counsel is advised that the inventor has previously filed papers requesting entry into the national phase of international application number PCT/EP2005/000221 and has appointed lawyers from another law firm to represent them before the USPTO. The US application number assigned to these papers is 10/522,451. The papers you filed on 11 July 2007 have been placed in application number 10/522,451. Since the inventors have previously appointed attorneys, no further communications with respect to this application will be mailed to you, unless a proper power of attorney is submitted to the PCT Legal Administration Office.

Any further correspondence with respect to this matter should be addressed to the Mail Stop PCT, Commissioner for Patents, Office of PCT Legal Administration, P.O. Box 1450, Alexandria, Virginia 22313-1450, with the contents of the letter marked to the attention of the Office of PCT Legal Administration.

Rafael Bacares
PCT Legal Examiner
PCT Legal Office
Tel: (571) 272-3276
Fax: (571) 273-0459

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A SUBMISSION UNDER 35 U.S.C. 371		ATTORNEY'S DOCKET NUMBER 37488-01300 U.S. APPLICATION NO. (If known, see 37 CFR 1.5) 11/795028 10/522, 451 PRIORITY DATE CLAIMED
INTERNATIONAL APPLICATION NO. PCT/EP/2005/000221	INTERNATIONAL FILING DATE January 12, 2005	
TITLE OF INVENTION Extrusion Coating Polyethylene		
APPLICANT(S) FOR DO/EO/US Laiho, Erkki; Sainio, Markku; Haapaniemi, Karoliina		
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:		
1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a submission under 35 U.S.C. 371. 2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a submission under 35 U.S.C. 371. 3. <input type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below. 4. <input type="checkbox"/> The US has been elected (Article 31). 5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)) a. <input checked="" type="checkbox"/> is attached hereto (required only if not communicated by the International Bureau). b. <input type="checkbox"/> has been communicated by the International Bureau. c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US). 6. <input type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)). a. <input type="checkbox"/> is attached hereto. b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4). 7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau). b. <input type="checkbox"/> have been communicated by the International Bureau. c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input type="checkbox"/> have not been made and will not be made. 8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). 9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). 10. <input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).		
Items 11 to 20 below concern document(s) or information included:		
11. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. 12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. 13. <input checked="" type="checkbox"/> A preliminary amendment. 14. <input type="checkbox"/> An Application Data Sheet under 37 CFR 1.76. 15. <input checked="" type="checkbox"/> A substitute specification. 16. <input type="checkbox"/> A power of attorney and/or change of address letter. 17. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 37 CFR 1.821- 1.825. 18. <input type="checkbox"/> A second copy of the published International Application under 35 U.S.C. 154(d)(4). 19. <input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).		

IAP8 Rec'd PCT/PTO 11 JUL 2007

10/522,451

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PTO-1390 (Rev. 09-2006)

Approved for use through 3/31/2007. OMB 0651-0021

U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

U.S. APPLICATION NO. (if known, see 37 CFR 1.51) Not Yet Assigned		INTERNATIONAL APPLICATION NO. PCT/EP/2005/000221		ATTORNEY'S DOCKET NUMBER 37488.01300	
20. Other items or information:					
The following fees have been submitted				CALCULATIONS PTO USE ONLY	
21. <input checked="" type="checkbox"/> Basic national fee (37 CFR 1.492(a))..... \$300				\$ 300.00	
22. <input checked="" type="checkbox"/> Examination fee (37 CFR 1.492(c))				\$ 200.00	
If the written opinion prepared by ISA/US or the international preliminary examination report prepared by IPEA/US indicates all claims satisfy provisions of PCT Article 33(1)-(4)..... \$0					
All other situations..... \$200					
23. <input checked="" type="checkbox"/> Search fee (37 CFR 1.492(b))				\$ 400.00	
If the written opinion of the ISA/US or the International preliminary examination report prepared by IPEA/US indicates all claims satisfy provisions of PCT Article 33(1)-(4)..... \$0					
Search fee (37 CFR 1.445(a)(2)) has been paid on the international application to the USPTO as an International Searching Authority..... \$100					
International Search Report prepared by an ISA other than the US and provided to the Office or previously communicated to the US by the IB..... \$400					
All other situations..... \$500					
TOTAL OF 21, 22 and 23 =				\$900.00	
<input type="checkbox"/> Additional fee for specification and drawings filed in paper over 100 sheets (excluding sequence listing in compliance with 37 CFR 1.821(c) or (e) in an electronic medium or computer program listing in an electronic medium) (37 CFR 1.492(j)). The fee is \$250 for each additional 50 sheets of paper or fraction thereof.					
Total Sheets	Extra Sheets	Number of each additional 50 or fraction thereof (round up to a whole number)	RATE		
- 100 =	/50 =		x \$250	\$	
Surcharge of \$130.00 for furnishing any of the search fee, examination fee, or the oath or declaration after the date of commencement of the national stage (37 CFR 1.492(h)).				\$	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$	
Total claims	40 - 20 =	20	x \$ 50	\$ 1000.00	
Independent claims	1 - 3 =	0	x \$200	\$	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$360	\$	
TOTAL OF ABOVE CALCULATIONS =				\$ 1900.00	
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. Fees above are reduced by 1/2.					
SUBTOTAL =				\$ 1900.00	
Processing fee of \$130.00 for furnishing the English translation later than 30 months from the earliest claimed priority date (37 CFR 1.492(i)).				\$	
TOTAL NATIONAL FEE =				\$ 1900	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				\$	
TOTAL FEES ENCLOSED =				\$ 1900	
				Amount to be refunded:	\$
				Amount to be charged	\$

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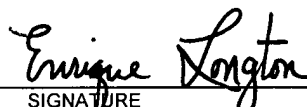
- a. ☒ A check in the amount of \$ 1900 to cover the above fees is enclosed.
- b. ☐ Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees.
A duplicate copy of this sheet is enclosed.
- c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 13-3250. A duplicate copy of this sheet is enclosed.
- d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card information should not be included on this form.** Provide credit card information and authorization on PTO-2038. The PTO-2038 should only be mailed or faxed to the USPTO. However, when paying the basic national fee, the PTO-2038 may NOT be faxed to the USPTO.

ADVISORY: If filing by EFS-Web, do NOT attach the PTO-2038 form as a PDF along with your EFS-Web submission. Please be advised that this is not recommended and by doing so your credit card information may be displayed via PAIR. To protect your information, it is recommended paying fees online by using the electronic payment method.

NOTE: Where an appropriate time limit under 37 CFR 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the International Application to pending status.

SEND ALL CORRESPONDENCE TO:

Customer No. 38647
Milbank, Tweed, Hadely & McCloy
1850 K St., NW Suite 1100
Washington, DC 20009



SIGNATURE

Enrique D. Longton

NAME

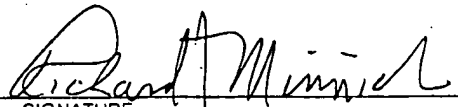
47,304

REGISTRATION NUMBER

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A SUBMISSION UNDER 35 U.S.C. 371		ATTORNEY'S DOCKET NUMBER KADZ 2000/3 U.S. APPLICATION NO. 107522451 (see 37 CFR 1.5)
INTERNATIONAL APPLICATION NO. PCT/EP05/000221	INTERNATIONAL FILING DATE 12 January 2005	PRIORITY DATE CLAIMED
TITLE OF INVENTION EXTRUSION COATING POLYETHYLENE		
APPLICANT(S) FOR DO/EO/US LAIHO, ERKKI et al.		
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:		
<ol style="list-style-type: none"> 1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a submission under 35 U.S.C. 371. 2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a submission under 35 U.S.C. 371. 3. <input type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below. 4. <input checked="" type="checkbox"/> The US has been elected (Article 31). 5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)) <ol style="list-style-type: none"> a. <input type="checkbox"/> is attached hereto (required only if not communicated by the International Bureau). b. <input type="checkbox"/> has been communicated by the International Bureau. c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US). 6. <input checked="" type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)). <ol style="list-style-type: none"> a. <input checked="" type="checkbox"/> is attached hereto. b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4). 7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) <ol style="list-style-type: none"> a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau). b. <input type="checkbox"/> have been communicated by the International Bureau. c. <input checked="" type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input type="checkbox"/> have not been made and will not be made. 8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). 9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). 10. <input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). 		
Items 11 to 20 below concern document(s) or information included:		
<ol style="list-style-type: none"> 11. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98, related PTO FORM 1449 & references 12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. 13. <input checked="" type="checkbox"/> A preliminary amendment. 14. <input type="checkbox"/> An Application Data Sheet under 37 CFR 1.76. 15. <input type="checkbox"/> A substitute specification. 16. <input type="checkbox"/> A power of attorney and/or change of address letter. 17. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 37 CFR 1.821- 1.825. 18. <input type="checkbox"/> A second copy of the published International Application under 35 U.S.C. 154(d)(4). 19. <input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4). 20. <input type="checkbox"/> Other items or information: 		

This collection of information is required by 37 CFR 1.414 and 1.491-1.492. The information is required to obtain or retain a benefit by the public, which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.11 and 1.14. This collection is estimated to take 15 minutes to complete, including gathering information, preparing, and submitting the completed form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Mail Stop PCT, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

U.S. APPLICATION NO. (if known, see 37 CFR 1.5)		INTERNATIONAL APPLICATION NO.		ATTORNEY'S DOCKET NUMBER	
10/522451		PCT/EP05/000221		KADZ 200043	
21. The following fees are submitted:				Applicant use	Office use only
<input checked="" type="checkbox"/> a) Basic national fee..... \$300.00				\$ 300.00	
<input checked="" type="checkbox"/> b) Examination fee..... \$200.00				\$ 200.00	
<input checked="" type="checkbox"/> c) Search fee..... \$500.00				\$ 500.00	
TOTAL OF ABOVE CALCULATIONS = \$1000.00				\$ 1,000.00	
<input type="checkbox"/> Additional fee for specification and drawings filed in paper, over 100 sheets (excluding sequence listing or computer program listing filed in an electronic medium). The fee is \$250 for each additional 50 sheets of paper or fraction thereof.					
Total Sheets	Extra sheets	Number of each additional 50 or fraction thereof (round up to a whole number)	RATE		
- 100 =	/50 =		x \$250.00	\$ 00.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$ 00.00	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	33 - 20 =	13	x \$50.00	\$ 650.00	
Independent claims	1 - 3 =	0	x \$200.00	\$ 00.00	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$360.00	\$ 00.00	
TOTAL OF ABOVE CALCULATIONS =				\$ 1,650.00	
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.				\$	
SUBTOTAL =				\$ 1,650.00	
Processing fee of \$130.00 for furnishing the English translation later than 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$	
TOTAL NATIONAL FEE =				\$ 1,650.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property.				\$	
TOTAL FEES ENCLOSED =				\$ 1,650.00	
Amount to be refunded:					\$
Amount to be charged:					\$
<p>a. <input checked="" type="checkbox"/> A check in the amount of \$ 1,650.00 to cover the above fees is enclosed.</p> <p>b. <input type="checkbox"/> Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.</p> <p>c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>06-0308</u>. A duplicate copy of this sheet is enclosed.</p> <p>d. <input type="checkbox"/> Fees are to be charged to a credit card. WARNING: Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.</p>					
NOTE: Where an appropriate time limit under 37 CFR 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the International Application to pending status.					
SEND ALL CORRESPONDENCE TO:					
Richard J. Minnich					
Fay, Sharpe, Fagan, Minnich & McKee, LLP					
1100 Superior Avenue, 7th Floor					
Cleveland, OH 44114					
				 SIGNATURE RICHARD J. MINNICH NAME 24,175 REGISTRATION NUMBER	

10/522451

DT01 Rec'd PCT/DTT 20 JAN 2005

PATENT

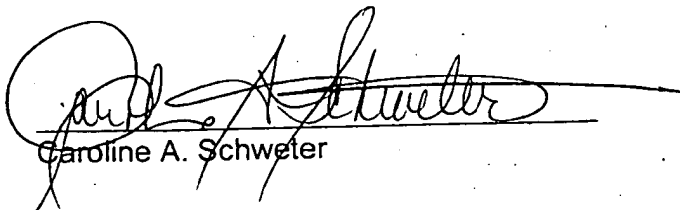
IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

INVENTOR(S) : ERKKI LAIHO et al.
TITLE : EXTRUSION COATING
: POLYETHYLENE
APPLICATION NO. : To be assigned
FILED : Herewith
ATTORNEY DOCKET NO. : KADZ 200043
Cleveland, OH 44114
January 20, 2005

CERTIFICATE OF EXPRESS MAILING UNDER 37 CFR 1.10

"Express Mail" Mailing Label Number: EV 494996906 US
Date of Deposit: January 20, 2005

I hereby certify that the attached **TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A SUBMISSION UNDER 35 U.S.C. 371** and accompanying documents are being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 C.F.R. 1.10 on the date indicated above and is addressed to: Commissioner For Patents, P.O. Box 1450, Alexandria, VA 22313-1450 on the date indicated above.


Caroline A. Schweter

PATENT APPLICATION SERIAL NO. _____

**U.S. DEPARTMENT OF COMMERCE
PATENT AND TRADEMARK OFFICE
FEE RECORD SHEET**

07/13/2007 GFREY1 00000046 11795028

01 FC:1631	300.00	DP
02 FC:1633	200.00	DP
03 FC:1642	400.00	DP
04 FC:1615	1000.00	DP

PTO-1556
(5/87)

71/795028

10/522,451

PATENT

Attorney Docket No. 37488.01300US

1AP8 Rec'd PCT/PTO 11 JUL 2007

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Application of:)
)
Laiho *et al.*)
) Group Art Unit: Not Yet Assigned
Application No: Not Yet Assigned)
Based on PCT/EP2005/000221)
) Examiner: Not Yet Assigned
Filed: January 12, 2005)
)
For: EXTRUSION COATING)
POLYETHYLENE)

Mail Stop Patent Application

Commissioner of Patents

P.O. Box 1450

Alexandria, VA 22313-1450

**PRELIMINARY AMENDMENT
UNDER 37 C.F.R. § 1.115**

Sir:

Prior to examination of this National Stage application of International Application No. PCT/EP2005/000221, please amend the above-identified application as follows:

Amendments to the **Specification** begin on page 2 of this paper.

Amendments to the **Claims** begin on page 3 of this paper.

The **Remarks** begin on page 11 of this paper.

IN THE SPECIFICATION

A clean copy of a substitute specification, as well as a separate, marked up version showing changes, are submitted herewith. The substitute specification contains no new matter.

LISTING OF THE CLAIMS

1. (Original) A multimodal polymer composition comprising
 - a. at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60000 g/mol;
 - b. at least one polyolefin (B) having a higher weight average molecular weight (M_w) than polymer (A); and
 - c. a filler (C), whereby
the polymer composition without filler (C) has a density of 940 kg/m³ or lower.
2. (Original) A polymer composition according to claim 1 characterized in that at least one polymer (A) is
 - (1) a polyolefin having a weight average molecular weight (M_w) of 10000 to 60000 g/mol, or
 - (2) a wax having weight average molecular weight (M_w) of less than 10000 g/mol.
3. (Amended) A polymer composition according to claim 2 1 characterized in that **at least one polymer (A) is**
 - (1) **a polyolefin having a weight average molecular weight (M_w) of 10000 to 60000 g/mol, or**
 - (2) **a wax having weight average molecular weight (M_w) of less than 10000 g/mol and**
the polyolefin (1) is a low density polyethylene (LDPE), a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE).
4. (Amended) A polymer composition according to claim ~~2 or 3~~ 1 characterized in that **at least one polymer (A) is**
 - (1) **a polyolefin having a weight average molecular weight (M_w) of 10000 to 60000 g/mol, or**
 - (2) **a wax having weight average molecular weight (M_w) of less than 10000 g/mol and**
the wax (2) is selected from one or more of

(2a) a polypropylene wax having weight average molecular weight (M_w) of less than 10000 g/mol or a polypropylene wax having weight average molecular weight (M_w) of less than 10000 g/mol, or

(2b) an alkyl ketene dimer wax having weight average molecular weight (M_w) of less than 10000 g/mol.

5. (Amended) A polymer composition according to ~~any one of the preceding claims 2 to 4~~ **claim 1** characterized in that the composition comprises a polyolefin (1) **having a weight average molecular weight (M_w) of 10000 to 60000 g/mol as polymer (A) and a wax (2) having weight average molecular weight (M_w) of less than 10000 g/mol** as a further polymer (A).
6. (Amended) A polymer composition according to ~~any one of the preceding claims 1 to 5~~ **claim 1** characterized in that the polymer (A) has a density of lower than 945 kg/m³.
7. (Amended) A polymer composition according to ~~any one of the preceding claims 1 to 6~~ **claim 1** characterized in that the multimodal polymer composition is at least a bimodal polymer composition.
8. (Amended) A polymer composition according to ~~any one of the preceding claims 1 to 7~~ **claim 1** characterized in that the polyolefin (B) has a weight average molecular weight (M_w) of higher than 80000 g/mol.
9. (Amended) A polymer composition according to ~~any one of the preceding claims 1 to 8~~ **claim 1** characterized in that the polyolefin (B) is a polyethylene.
10. (Amended) A polymer composition according to ~~claim 9~~ **1** characterized in that the polyolefin (B) is a low density polyethylene (LDPE), a linear low density polyethylene (LLDPE) or linear medium density polyethylene (LMDPE).
11. (Amended) A polymer composition according to ~~any one of the preceding claims 1 to 10~~ **claim 1** characterized in that the total polymer composition comprises 1 to 50 wt% of polymer (A), 40 to 90 wt% of polyolefin (B) and 1 to 50 wt% of filler (C).

12. (Amended) A polymer composition according to ~~any one of the preceding claims 1 to 11~~ claim 1 characterized in that the polymer composition without filler (C) has melt flow rate MFR₂, according to ISO 1133, at 190 °C, of 5 to 20 g/10min.
13. (Amended) A polymer composition according to ~~any one of the preceding claims 1 to 12~~ claim 1 characterized in that the polymer composition without filler (C) has melt flow rate MFR₅, according to ISO 1133, at 190 °C, of 20 to 40 g/10min.
14. (Amended) A polymer composition according to ~~any one of the preceding claims 1 to 13~~ claim 1 characterized in that the polymer composition without filler (C) has melt flow ratio MFR₅/MFR₂ of 2.5 to 4.5.
15. (Amended) A polymer composition according to ~~any one of the preceding claims 1 to 14~~ claim 1 characterized in that the polymer composition without filler (C) has a ratio of the weight average molecular weight (M_w) to the number average molecular weight (M_n) from 8 to 25.
16. (Amended) A polymer composition according to ~~any one of the preceding claims 1 to 15~~ claim 1 characterized in that 95 wt% of the filler (C) has a particle size of less than 10 µm.
17. (Amended) A polymer composition according to ~~any one of the preceding claims 1 to 16~~ claim 1 characterized in that the filler (C) is talc.
18. (Amended) A polymer composition according to ~~any one of the preceding claims 1 to 17~~ claim 1 characterized in that the polymer composition comprises additionally antioxidants(s) and/or process stabilizers of less than 2000 ppm in the total composition.
19. (Amended) A polymer composition according to ~~any one of the preceding claims 1 to 18~~ claim 1 characterized in that the polymer composition is a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE), whereby polymer (A) and polyolefin (B) are produced in a multi step polymerization process.

20. (Amended) A polymer composition according to ~~any one of the preceding claims 1 to 19~~ claim 1 characterized in that the polymer composition is a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE), whereby polymer (A) and polyolefin (B) are produced in a multi step polymerization process and the amount of comonomer units in a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE) is 0.1 to 1.0 mol %.
21. (Amended) A polymer composition according to ~~claim 19 or 20~~ 1 characterized in that the polymer ~~(A) and the polyolefin (B)~~ composition is a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE), whereby polymer (A) and polyolefin (B) are produced in a multi step polymerization process and the polymer (A) and the polyolefin (B) is a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE) and the comonomer units are selected from the group consisting of C₃ α -olefin, C₄ α -olefin, C₅ α -olefin, C₆ α -olefin, C₇ α -olefin, C₈ α -olefin, C₉ α -olefin, C₁₀ α -olefin, C₁₁ α -olefin, C₁₂ α -olefin, C₁₃ α -olefin, C₁₄ α -olefin, C₁₅ α -olefin, C₁₆ α -olefin, C₁₇ α -olefin, C₁₈ α -olefin, C₁₉ α -olefin and C₂₀ α -olefin.
22. (Amended) A polymer composition according to ~~any one of the preceding claims 1 to 18~~ claim 1 characterized in that the polymer (A) is a wax (2) according to ~~claim 4~~ which is selected from one or more of
(2a) a polypropylene wax having weight average molecular weight (M_w) of less than 10000 g/mol or a polypropylene wax having weight average molecular weight (M_w) of less than 10000 g/mol, or
(2b) an alkyl ketene dimer wax having weight average molecular weight (M_w) of less than 10000 g/mol and
the polyolefin (B) is a linear low density polyethylene (LLDPE) or low density polyethylene (LDPE).
23. (Amended) A polymer composition according to ~~claim 22~~ 1 characterized in that the polymer (A) is a wax (2) which is selected from one or more of

(2a) a polypropylene wax having weight average molecular weight (M_w) of less than 10000 g/mol or a polypropylene wax having weight average molecular weight (M_w) of less than 10000 g/mol, or

(2b) an alkyl ketene dimer wax having weight average molecular weight (M_w) of less than 10000 g/mol,

the polyolefin (B) is a linear low density polyethylene (LLDPE) or low density polyethylene (LDPE) and the polymer composition comprises additionally a polyolefin (1) being a linear low density polyethylene (LLDPE) as a further polymer (A).

24. (Amended) A polymer composition according to claim 20 ~~or 21~~ 1 characterized in that the polymer composition is a linear low density polyethylene (LLDPE) whereby polymer (A) and polyolefin (B) are produced in a multi step polymerization process, the amount of comonomer units in a linear low density polyethylene (LLDPE) is 0.1 to 1.0 mol % and polyolefin (1) (polymer (A)) being a linear low density polyethylene (LLDPE) is the lower molecular weight fraction of LLDPE and polyolefin (B) being a linear low density polyethylene (LLDPE) is the higher molecular weight fraction of the LLDPE.
25. (Amended) A polymer composition according to claim 24 1 characterized in that the polymer composition is a linear low density polyethylene (LLDPE) whereby polymer (A) and polyolefin (B) are produced in a multi step polymerization process, the amount of comonomer units in a linear low density polyethylene (LLDPE) is 0.1 to 1.0 mol % and polyolefin (1) (polymer (A)) being a linear low density polyethylene (LLDPE) is the lower molecular weight fraction of LLDPE and polyolefin (B) being a linear low density polyethylene (LLDPE) is the higher molecular weight fraction of the LLDPE and the polymer (A) and polyolefin (B) are a mechanical blend, preferably an in-situ blend produced in a multistage polymerization process.
26. (Amended) A multi-layer material comprising
- a. a substrate as a first layer (I)

- b. a multimodal polymer composition according to ~~any one of the preceding claims~~ claim 1 as at least a further layer (II).
27. (Original) A multi-layer material according to claim 26 characterized in that the substrate is selected from the group consisting of paper, paperboard, aluminium film and plastic film.
28. (Amended) A multi-layer material according to claim 26 ~~or 27~~ characterized in that the multi-layer material comprises as a further layer (III) comprising a low density polyethylene (LDPE).
29. (Amended) A multi-layer material according to ~~any one of the preceding claims 26 to 28~~ claim 26 characterized in that the low density polyethylene (LDPE) layer (III) has a melt flow rate MFR₂, according to ISO 1133, at 190°C, of at least 5 g/10 min.
30. (Amended) A film comprising a multimodal polymer composition according to ~~any one of the preceding claims 1 to 25~~ claim 1.
31. (Amended) A process for producing a composition according to ~~any one of the preceding claims 1 to 25~~ claim 1 characterized in that
- a. polymer (A) and polyolefin (B) are produced together in a multistage process comprising a loop reactor and a gas phase reactor, wherein polymer (A) is generated in at least one loop reactor and the polyolefin (B) is generated in a gas phase reactor; and
- b. filler (C) and the composition comprising polymer (A) and polyolefin (B) are blended together and compounded.
32. (Original) A process for producing a composition according to claim 31 characterized in that the catalyst used for the process producing the composition comprising polymer (A) and polyolefin (B) is a high activity procatalyst comprising a particulate inorganic support, a chlorine compound deposited on the support, wherein the chlorine compound is the same as or different from the titanium compound, whereby the inorganic support is contacted with an alkyl metal chloride which is soluble in non-polar hydrocarbon solvents, and has the formula $R_nMECL_{3-n})_m$

wherein R is a C₁-C₂₀ alkyl group, Me is a metal of group III(13) of the periodic table, n=1 or 2 and m=1 or 2, to give a first reaction product, and the first reaction product is contacted with a compound containing hydrocarbyl and hydrocarbyl oxide linked to magnesium which is soluble in non-polar hydrocarbon solvents, to give a second reaction product, and the first reaction product is contacted with a compound containing hydrocarbyl and hydrocarbyl oxide linked to magnesium which is soluble in non-polar hydrocarbon solvents, to give a second reaction product, and the second reaction product is contacted with a titanium compound which contains chlorine, having the formula Cl_xTi(OR^{IV})_{4-x} wherein R^{IV} is a C₂-C₂₀ hydrocarbyl group and x is 3 or 4, to give the procatalyst.

33. (Amended) A process for producing a multi-layer material according to ~~any one of the preceding claims 26 to 29~~ **claim 26** characterized in that the multimodal polymer composition ~~according to any one of claims 1 to 25~~ **comprising**
- a. **at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60000 g/mol;**
 - b. **at least one polyolefin (B) having a higher weight average molecular weight (M_w) than polymer (A); and**
 - c. **a filler (C), whereby**
the polymer composition without filler (C) has a density of 940 kg/m³ or lower is applied on the substrate by a film coating line comprising an unwind, a wind, a chill roll and a coating die.
34. (Amended) Use of the multimodal polymer composition according to ~~any one of the preceding claims 1 to 25~~ **claim 1** for extrusion coating.
35. (Amended) Use according to claim 34 characterized in that the ~~polymer extrusion composition according to any one of the preceding claims 1 to 25 is used for extrusion coating producing a multi-layer material according to any one of the claim 26 to 29~~ **multimodal polymer composition is used for extrusion coating producing a multi-layer material comprising**
- a. **a substrate as a first layer (I)**

b. **a multimodal polymer composition according to claim 1 as at least a further layer (II).**

36. (Amended) Use of the multimodal polymer composition according to ~~any one of the preceding claims 1 to 25~~ **claim 1** for a film, preferably for a cast film.
37. (New) A polymer composition according to claim 1 characterized in that the composition comprises a polyolefin (1) having a weight average molecular weight (M_w) of 10000 to 60000 g/mol as polymer (A) and a wax (2) having weight average molecular weight (M_w) of less than 10000 g/mol as a further polymer (A) wherein the polyolefin (1) is a low density polyethylene (LDPE), a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE).
38. (New) A polymer composition according to claim 1 characterized in that the composition comprises a polyolefin (1) having a weight average molecular weight (M_w) of 10000 to 60000 g/mol as polymer (A) and a wax (2) having weight average molecular weight (M_w) of less than 10000 g/mol as a further polymer (A) wherein the wax (2) is selected from one or more of
(2a) a polypropylene wax having weight average molecular weight (M_w) of less than 10000 g/mol or a polypropylene wax having weight average molecular weight (M_w) of less than 10000 g/mol, or
(2b) an alkyl ketene dimer wax having weight average molecular weight (M_w) of less than 10000 g/mol.
39. (New) A polymer composition according to claim 1 characterized in that the polymer composition is a linear low density polyethylene (LLDPE) whereby polymer (A) and polyolefin (B) are produced in a multi step polymerization process, the comonomer units are selected from the group consisting of C₃ α -olefin, C₄ α -olefin, C₅ α -olefin, C₆ α -olefin, C₇ α -olefin, C₈ α -olefin, C₉ α -olefin, C₁₀ α -olefin, C₁₁ α -olefin, C₁₂ α -olefin, C₁₃ α -olefin, C₁₄ α -olefin, C₁₅ α -olefin, C₁₆ α -olefin, C₁₇ α -olefin, C₁₈ α -olefin, C₁₉ α -olefin and C₂₀ α -olefin and polyolefin (1) (polymer (A)) being a linear low density polyethylene (LLDPE) is the lower molecular weight fraction of LLDPE and polyolefin (B)

being a linear low density polyethylene (LLDPE) is the higher molecular weight fraction of the LLDPE.

40. (New) A polymer composition according to claim 1 characterized in that the polymer composition is a linear low density polyethylene (LLDPE) whereby polymer (A) and polyolefin (B) are produced in a multi step polymerization process, the comonomer units are selected from the group consisting of C₃ α -olefin, C₄ α -olefin, C₅ α -olefin, C₆ α -olefin, C₇ α -olefin, C₈ α -olefin, C₉ α -olefin, C₁₀ α -olefin, C₁₁ α -olefin, C₁₂ α -olefin, C₁₃ α -olefin, C₁₄ α -olefin, C₁₅ α -olefin, C₁₆ α -olefin, C₁₇ α -olefin, C₁₈ α -olefin, C₁₉ α -olefin and C₂₀ α -olefin and polyolefin (1) (polymer(A)) being a linear low density polyethylene (LLDPE) is the lower molecular weight fraction of LLDPE and polyolefin (B) being a linear low density polyethylene (LLDPE) is the higher molecular weight fraction of the LLDPE and the polymer (A) and polyolefin (B) are a mechanical blend, preferably an in-situ blend produced in a multistage polymerization process.

REMARKS

Amendments to the Specification

The specification has been amended to correct for typographical errors in the specification. The amendments also conform the original specification to requirements according to U.S. practice. A substitute specification is being submitted herewith. The amendments and substitute specification contain no new material.

Amendments to the Claims

The claims have been amended to correct for typographical errors and to conform the claims according to U.S. practice, which does not allow for multiple dependent claims. Support for the amendments can be found directly in the claims. For example, support for amendments in claims 3 and 4 can be found in claim 2. Support for amendments to claims 5 and 19 can be found in claim 4. Support for amendments to claims 20 and 21 can be found in claim 19. Support for amendments in claims 23, 24, and 25 can be found in claims 22, 20, and 24, respectively. Likewise, support for amendments to claim 33 and 34 can be found in claims 1 and 26, respectively.

New claims 37-40 have been added. These new claims also account for the multiple dependencies in the PCT application. Support for amendments to claims 37 and 38 can be found in claims 2 and 3 and 2 and 4, respectively. Support for amendments to claims 37 and 38 can be found in claims 2 and 3 and 2 and 4, respectively. Finally, support for claims 39 and 40 can be found in claims 19, 21 and 24, and claims 19, 21 and 25, respectively.

No new matter is added by these amendments.


Entry of the foregoing amendments is respectfully requested. No prohibited new matter is introduced by these amendments.

EXCEPT for issue fees payable under 37 C.F.R. § 1.18, the Director is hereby authorized by this paper to charge any additional fees during the entire pendency of this application, including fees due under 37 C.F.R. §§ 1.16 and 1.17 which may be required, including any required extension of time fees, or credit any overpayment to

Deposit Account No. 13-3250, reference No. 37488.01100US. This paragraph is intended to be a **CONSTRUCTIVE PETITION FOR EXTENSION OF TIME** in accordance with 37 C.F. R. § 1.136(a)(3).

Respectfully submitted,

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EXTRUSION COATING POLYETHYLENE**FIELD OF THE INVENTION**

[0001] The present invention relates to a polymer composition suitable for extrusion coating and films, preferably cast films having good chemical properties and barrier properties, in particular, a low water-vapor transmission rate (WVTR) and a low curling. Additionally, the present invention relates to the process for producing the inventive composition and its use. Moreover, the present invention is related to a multi-layer material comprising the polymer composition as well as to a process of said multi-layer material.

BACKGROUND OF THE INVENTION

[0002] One of the largest and most rapidly growing polyolefin-processing method is extrusion coating. The largest single volume of coated materials are different papers and paperboards, which are used for a variety of packaging applications. Other material frequently coated are polymer films, cellophane, aluminum foil, freezer wrap paper and fabrics of various kinds. One target for the improvement of coated articles is to reduce the water-vapor transmission rate (WVTR) as much as possible. A coated material with a low water-vapor transmission rate (WVTR) can for example protect the products wrapped therein much better. The demanded requirement applies, of course, not only to coated materials but also to cast films used for packaging or containers. In both cases, a low water-vapor transmission rate is required. Much effort has been undertaken to improve the water-vapor transmission rate of coated materials as well as for cast films. To date, several new polymer compositions have been developed and much effort has been undertaken to find appropriate fillers to improve the barrier properties significantly. Furthermore, different polymers have been designed as cyclo-olefin copolymers (COC) and liquid crystal polymers (LCP). However, these materials have the drawback of being expensive and having minor processability properties.

[0003] WO 00/71615 discloses for example the use of a bimodal high density polyethylene (HDPE) with a melt flow rate, MFR₂, of 5 g/10 min and a density of 957 kg/m³ for extrusion coating. No information is given how to improve the water-vapor transmission rate (WVTR).

[0004] WO 00/34580 describes release liner for pressure-sensitive adhesive labels. The release-liner contains a paper wrap, a filled polymer layer, and, on the opposite of the paper web, an extrudate, e.g. polyethylene, and on the top of the extrudate, a release film. The filled polymer layer can be polyethylene and the filler is an inert particulate, such as silica, mica, clay, talc and titanium oxide. The filler is present in 15 to 40 wt% of the composition.

[0005] US 4,978,572 describes a laminated film having three layers. The first layer comprises a thermoplastic resin and 0.3 to 30 wt% white inorganic particles. The second layer comprises an ethylene copolymer, 0.5 to 90 wt% of a substance giving anti-block action and anti-oxidant. The third layer comprises a metallized thermoplastic. The substance giving anti-block action of the second layer may be silica or talc. The laminated film is reported to have good mechanical strength and good barrier properties.

[0006] Even though the prior art offers already a variety of products having good water-vapor transmission rates (WVTR), there is still demand for a significant improvement of these properties. One significant disadvantage in polymer compositions comprising fillers reducing the water-vapor transmission rate (WVTR) is the low dispersion of the fillers incorporated in the polymer matrix. Conventional mechanical incorporation frequently results in poor dispersion as usual fillers form multi-layer aggregation caused by incompatibility with polymer matrix. One consequence of the described phenomenon is that the water-vapor transmission rate (WVTR) varies considerably in the layer leading to unsatisfying average values for the WVTR. Secondly, the low dispersion of the filler causes an easy upcurling of the polymer composition coated on the materials. Hence, a uniform dispersion of fillers incorporated in a polymer composition should improve the water-vapor transmission rate significantly, and, additionally, the curling properties of a coated material should be enhanced.

[0007] Hence, the object of the present invention is to improve the water-vapor transmission rate (WVTR).

[0008] The present invention is based on the finding that the object can be addressed by a polymer composition comprising a polymer having a low average molecular weight enabling an enhanced and uniform dispersion of fillers incorporated in the polymer composition.

SUMMARY OF THE INVENTION

[0009] The present invention therefore provides a multimodal polymer composition comprising at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60,000 g/mol; at least one polyolefin (B) having a higher weight average molecular weight (M_w) than polymer (A); and a filler (C) whereby the polymer composition without filler (C) has a density of 940 kg/m³ or lower.

[00010] It is preferred that the polymer composition consists of at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60,000 g/mol; at least one polyolefin (B) having a higher weight average molecular weight (M_w) than polymer (A); and filler (C) whereby the polymer composition without filler (C) has a density of 940 kg/m³ or lower.

[00011] Accordingly, the polymer composition according to this invention is multimodal with respect to the molecular weight distribution. “Multi-modal” or “multimodal distribution” describes a frequency distribution that has several relative maxima. In particular, the expression “modality of a polymer” refers to the form of its molecular weight distribution (MWD) curve, i.e. the appearance of the graph of the polymer weight fraction as a function of its molecular weight. The molecular weight distribution (MWD) of a polymer produced in a single polymerization stage using a single monomer mixture, a single polymerization catalyst and a single set of process conditions (i.e. temperature, pressure, etc.) shows a single maximum the breadth of which depends on catalyst choice, reactor choice, process conditions, etc., i.e. such a polymer is monomodal.

[00012] This inventive composition is characterized by a very low water-vapor transmission rate (WVTR) and also by low curling-values for extrusion-coated layers. These improved properties are reached by a much better dispersion of the filler (C) in the polymer mixture of polymer (A) and polyolefin (B) compared with an unmimodal polymer having the same melt index and density for both extrusion-coated layers and cast films.

[00013] Hence, the polymer composition according to this invention is a multimodal polymer including bimodal polymer composition consisting of at least two different polymers

having two different molecular weight distribution curves and are blended mechanically or *in situ* during the preparation thereof. Preferably the polymer composition is at least a bimodal mechanical or *in-situ* blend of a polyolefin (1) (as polymer (A)) and polymer (B). In case such a bimodal blend comprises further a wax (2) as an additional polymer (A), then the final polymer composition may also be trimodal.

[00014] The molecular weight distribution (MWD) is the relation between the numbers or molecules in a polymer and their individual chain length. The molecular weight distribution (MWD) is often given as a number, which normally means weight average molecular weight (M_w) divided by number average molecular weight (M_n).

[00015] The weight average molecular weight (M_w) is the first moment of a plot of the weight of polymers in each molecular weight range against molecular weight. In turn, the number average molecular weight (M_n) is an average molecular weight of a polymer expressed as the first moment of a plot of the number of molecules in each molecular weight range against the molecular weight. In effect, this is the total molecular weight of all molecules divided by the number of molecules.

[00016] The number average molecular weight (M_n) and the weight average molecular weight (M_w) as well as the molecular weight distribution (MWD) are determined according to ISO 16014.

[00017] The weight average molecular weight (M_w) is a parameter for the length of the molecules in average. Low M_w -values indicate that the chain length of the molecules is rather short in average. It has been found out that a polymer mixture comprising a polymer (A) with M_w -values of lower than 60,000 g/mol contributes inter alia to better barrier properties and better dispersion of the filler (C). Such better dispersion improves the water-vapor transmission rate (WVTR) as well as the curling resistance positively.

[00018] Hence, as a further requirement of the present invention, the multimodal polymer composition must comprise at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60,000 g/mol. It is in particular preferred that at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60,000 g/mol is at least one

polyolefin (1) having a weight average molecular weight (M_w) of 10,000 to 60,000 g/mol, more preferably of 20,000 to 50,000 g/mol and/or at least one wax (2) having a weight average molecular weight (M_w) of less than 10,000 g/mol, more preferably in the range of 500 to 10,000 g/mol.

[00019] Moreover, it is preferred that the polyolefin (1) is a polyethylene or polypropylene, more preferably a polyethylene. The polyolefin (1) can be a homopolymer or copolymer. It is preferred that the polyolefin (1) is a homopolymer or copolymer of propylene or ethylene, more preferred the polyolefin (1) is a homopolymer or copolymer of ethylene. Most preferably the polyolefin (1) is a low density polyethylene (LDPE), a linear low density polyethylene (LLDPE), or a linear medium density polyethylene (LMDPE). LDPE, LLDPE and LMDPE are equally suitable alternatives for polyolefin (1), e.g. where a LLDPE or a LMDPE is applicable also a LDPE can be used and vice versa.

[00020] In case polymer (A) is a wax (2), it is preferred that it is selected from one or more of a polypropylene wax (2a) having a weight average molecular weight (M_w) of less than 10,000 g/mol, more preferably in the range of 500 to 10,000 g/mol, still more preferably in the range of 1,000 to 9,000 g/mol, yet more preferably in the range of 2,000 to 8,000 g/mol and most preferably in the range of 4,000 to 8,000 g/mol or a polyethylene wax having a weight average molecular weight (M_w) of less than 10,000 g/mol, more preferably in the range of 500 to 10,000 g/mol, still more preferably in the range of 1,000 to 9,000 g/mol, yet more preferably in the range of 2,000 to 8,000 g/mol and most preferably in the range of 4,000 to 8,000 g/mol, and an alkyl ketene dimer wax having weight average molecular weight (M_w) of less than 10,000 g/mol more preferably lower than 5,000 g/mol yet more preferably lower than 1,000 g/mol. In turn the alkyl ketene dimer wax has preferably weight average molecular weight (M_w) of at least 100 g/mol. Most preferred the alkyl ketene dimer wax (2b) has weight average molecular weight (M_w) in the range of 250 to 1,000 g/mol.

[00021] The terms “at least one polymer (A)”, “at least one polyolefin (1)”, or “at least one wax (2)” shall indicate that more than one polymer (A), polyolefin (1) or wax (2) can be present in the multimodal polymer composition. It is preferred that three, two or one different polymers (A) as defined above are used in a multimodal polymer composition. Still more

preferred is that wax (2), preferably a polypropylene wax (2a) or an alkyl ketene dimer wax (2b) as defined above is used as a component (A) only. In case the component (A) comprises a polyolefin (1) as defined above, it is preferred that a wax (2) is present in the multimodal polymer composition as a further polymer (A). In such cases the multimodal composition is preferably trimodal comprising polyolefin (1), wax (2) and polyolefin (B) having different centered maxima in their molecular weight distribution, e.g. having different weight average molecular weights (M_w). The use of the wax (2) has the benefit that the amorphous region of the polymer matrix, which may be a mix of polyolefin (1) and polyolefin (B), is filled up and improves thereby the barrier properties.

[00022] It is preferred that not only the final polymer composition has a specific density of 940 kg/m^3 or lower but also the polymer (A) shall have a density of lower than 945 kg/m^3 . It is preferred that polyolefin (1) when used as polymer (A) has a density lower than 945 kg/m^3 , more preferably is in a range of 905 to 935 kg/m^3 , still more preferably in the range of 910 to 930 kg/m^3 and most preferred in the range of 915 to 925 kg/m^3 . Preferably, the polyolefin (1) is a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE). In turn, also a low density polyethylene (LDPE) produced in a high pressure process by free radical polymerization is applicable as a polyolefin (1). The linear low density polyethylene (LLDPE) or the linear medium density polyethylene (LMDPE) is produced by a process as described for the polyolefin (B) below.

[00023] The molecular weight distribution (MWD) of the polymer composition is further characterized by the way of its melt flow rate (MFR) according to ISO 1133 at 190°C . The melt flow rate (MFR) mainly depends on the average molecular weight. The reason for this is that long molecules give the material a lower flow tendency than short molecules.

[00024] An increase in molecular weight means a decrease in the MFR-value. The melt flow rate (MFR) is measured in g/10 min of the polymer discharged under specific temperature and pressure conditions and is the measure of a viscosity of the polymer which in turn for each type of polymer is mainly influenced by its molecular weight distribution, but also by its degree of branching. The melt flow rate measured under a load of 2.16 kg (ISO 1133) is denoted as MFR2. In turn, the melt flow rate measured with 5 kg load (ISO 1133) is denoted as MFR5.

[00025] In case polymer (A) is a polyolefin (1), it is preferred that MFR₂ is in the range of 1.0 to 20.0 g/10 min and more preferably in the range of 2.0 to 15.0 g/10 min and for example in the range of 3.0 to 10.0 g/10 min. According to one embodiment, the polyolefin (1) is a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE) with MFR₂ as given above. In turn, polyolefin (1) can also be a low density polyethylene (LDPE) having a density in the range as stated in this paragraph. The low density polyethylene (LDPE) is produced in a high-pressure process by free radical polymerization. In turn, the linear low density polyethylene (LLDPE) or the linear medium density polyethylene (LMDPE) is produced as described for polyolefin (B).

[00026] In case polymer (A) is an ethylene homopolymer, it is preferred that the ethylene homopolymer contains less than 0.2 mol%, more preferably less than 0.1 mol% and most preferably less than 0.05 mol% units derived from alpha-olefins other than ethylene. It is in particular preferred that the polymer (A) is an ethylene co-polymer, more preferably having a weight average molecular weight from 10,000 to 60,000 g/mol, still more preferably from 20,000 to 50,000 g/mol. Yet more preferably the polymer (A) is an ethylene copolymer having a density of 905 to 935 kg/m³, more preferably of 910 to 930 kg/m³, most preferably of 915 to 925 kg/m³ and having a melt index MFR₂ from 1.0 to 20.0 g/10 min, more preferably from 2.0 to 15.0 g/10 min, most preferably of 3.0 to 10.0 g/10 min. Preferably, the ethylene copolymer comprises, more preferably consists of, comonomer units as defined below for the LLDPE. It is in particular preferred that the ethylene copolymer fulfills all properties as stated above simultaneously.

[00027] In case polymer (A) is a wax (2a), namely a polypropylene wax or a polyethylene wax, it is preferred that the wax (2a) has a weight average molecular weight (M_w) in the range of 500 to 10,000 g/mol, more preferably in the range of 1,000 to 9,000 g/mol, still more preferably in the range of 2,000 to 8,000 g/mol and most preferably in the range of 4,000 to 8,000 g/mol. Further preferred ranges for the weight average molecular weight (M_w) of the wax (2a), in particular the polypropylene or polyethylene wax, is in the range of 4,000 to 7,000 g/mol, still more preferably in the range of 5,000 to 6,000 g/mol and most preferably in the range of 5,300 to 5,400 g/mol. Additionally, it is preferred that the wax (2a), in particular the polypropylene wax or polyethylene wax, has a z-average molecular weight of 9,100 to 40,000 g/mol, more

preferably from 500 to 20,000 g/mol and most preferably from 10,000 to 12,000 g/mol. It is additionally preferred that the wax (2a), in particular the polypropylene wax or the polyethylene wax, has a number average molecular weight (M_n) of 100 to 20,000 g/mol, more preferably of 500 to 3,000 g/mol.

[00028] Moreover, it is preferred that wax (2a), in particular polypropylene wax or polyethylene wax, has a specific molecular weight distribution (MWD) which is the relation between the number of molecules in the polymer and their individual chain length. The molecular weight distribution is given as a number which means weight average molecular weight divided by number average molecular weight (M_w/M_n). It is preferred that the wax (2a), in particular the polypropylene wax or the polyethylene wax, has an MWD in the range of 1 to 5, more preferably in the range of 1.5 to 4.

[00029] In addition, it is preferred that the wax (2a), in particular the polypropylene wax or the polyethylene wax, has a melting temperature in DSC-analysis of below 150°C, more preferably below 140°C, still more preferably in the range of 95 to 130°C, most preferably in a range of 105 to 115°C.

[00030] In case a wax (2b), namely an alkyl-ketene dimer, is employed as polymer (A), it is preferred that the weight average molecular weight (M_w) of the wax (2b) is higher than 100 g/mol. In turn, it is preferred that the weight average molecular weight of the wax (2b) is lower than 10,000 g/mol, more preferably lower than 5,000 g/mol, still more preferably lower than 1,000 g/mol. Preferred ranges for the weight average molecular weight (M_w) of the wax (2b) is 100 to 10,000 g/mol, more preferably 250 to 1,000 g/mol. Additionally, it is preferred that the wax (2b) has a number average molecular weight (M_n) of 100 to 20,000 g/mol, more preferably in the range of 100 to 800 g/mol. In addition, it is preferred that wax (2b) has a melting temperature in DSC-analysis below 140°C, more preferably below 100°C. A preferred range for the melting temperature in DSC-analysis is 50 to 90°C, more preferably 50 to 70°C.

[00031] As a further requirement, according to the present invention, the polyolefin (B) shall have a higher weight average molecular weight (M_w) than polymer (A). It is preferred that the polyolefin (B) has a weight average molecular weight (M_w) of higher than 80,000 g/mol, more preferably higher than 100,000 g/mol. The upper limit for the weight average molecular

weight for polyolefin (B) shall preferably not be higher than 300,000 g/mol, more preferably not higher than 200,000 g/mol. The preferred range for the weight average molecular weight for polyolefin (B) is 80,000 to 300,000 g/mol, more preferably from 100,000 to 200,000 g/mol. Preferably, polyolefin (B) is a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE), which has been preferably produced in a low medium pressure process in the presence of a polymerization catalyst (i.e. a Ziegler-Natta catalyst or a metallocene catalyst). A linear low density polyethylene (LLDPE) and a linear medium density polyethylene (LMDPE) has a density lower than 945 kg/m³, more preferably in the range of 905 to 935 kg/m³, still more preferably in the range of 910 to 930 kg/m³ and most preferred in the range of 915 to 925 kg/m³. However, also a low density polyethylene (LDPE) is also applicable for the polyolefin (B). A low density polyethylene (LDPE) has the same density ranges as the LLDPE or the LMDPE as stated in this paragraph and is a product from a high pressure polymerization process characterized by a highly branched chain structure. LDPE, LLDPE and LMDPE are equally suitable alternatives for polyolefin (B), e.g. where a LLDPE or a LMDPE is applicable also a LDPE can be used and vice versa.

[00032] According to this invention, more than one polyolefin (B) can be used. Accordingly, the invention also comprises the possibility of any mixture of a linear low density polyethylene (LLDPE), a linear medium density polyethylene (LMDPE) and a low density polyethylene (LDPE).

[00033] The MFR₂ of the polyolefin (B) is preferably in the range of 1.0 to 20.0 g/10 min, more preferably in the range of 2.0 to 15.0 g/10 min, in the range of e.g. 3.0 to 10.0 g/10 min. It is in particular preferred that the linear low density polyethylene (LLDPE) and the linear medium density polyethylene (LMDPE) have such melt flow characteristics. In turn, also the low density polyethylene (LDPE) suitable as a polyolefin (B) may have the melt flow characteristics as given in this paragraph.

[00034] It is preferred that the polyolefin (B) is a polyethylene. In case the polyolefin (B) is a polyethylene, it may be an ethylene homopolymer or an ethylene copolymer. In case for the polyolefin (B) an ethylene homopolymer is employed, then preferably an ethylene homopolymer is used as defined for polymer (A). In case an ethylene copolymer is employed

for polyolefin (B), then preferably an ethylene copolymer is used as defined below. It is in particular preferred that polyolefin (B) is a low density polyethylene (LDPE), a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE).

[00035] According to one embodiment, the polymer composition according to this invention is a linear low density polyethylene (LLDPE) comprising polyolefin (1) (polymer (A)) as a low molecular weight fraction of LLDPE and polyolefin (B) as a high molecular weight fraction of LLDPE. This linear low density polyethylene (LLDPE) may be a mechanical blend, preferably an in-situ blend produced in a multi-stage process. Preferably said composition comprises wax (2) as a further polymer (A).

[00036] It is preferred that the polymer composition as defined above comprises 1 to 50 wt% of polymer (A), 40 to 90 wt% of polyolefin (B) and 1 to 50 wt% of filler (C), more preferably of 5 to 40 wt%, and most preferably of 10 to 35 wt%. In case the polymer composition is produced in an *in situ* polymerization process, e.g. a sequential step process by utilizing reactors coupled in series and described as above, it is preferred that the polymer (A) may range from 40 to 60 wt%, more preferably 49 to 55 wt% in the polymer mix without filler (C). In turn, it is preferred that in such a polymer mix, the polyolefin (B) ranges from 60 to 40 wt%, more preferably from 51 to 45 wt%. Preferably, the total polymer composition comprises 50 to 99 wt% of said polymer mix and of 1 to 50 wt% filler (C), more preferably of 5 to 40 wt%, and most preferably of 10 to 35 wt%.

[00037] In case polymer (A) and polyolefin (B) are blended mechanically, it is preferred that polymer (A) ranges from 1 to 30 wt% and, more preferably, from 1 to 20 wt% in the total polymer composition. These ranges apply in particular in case for polymer (A) a wax (2) is used only.

[00038] The last requirement according to the present invention is that the multimodal polymer composition additionally comprises a filler (C). Any filler having a positive influence on the water-vapor transmission rate (WVTR) can be used. Preferably, the filler shall be lamellar, such as clay, mica or talc. More preferably, the filler shall be finely divided. The finely divided filler consists of about 95 wt% of particles having particle sizes of less than 10 μm , and about 20-30 wt% of particles having a particle size of less than 1 μm . In the present

invention all layer materials may be used as long as they have the ability to disperse in the polymer composition. The filler may either be a clay-based compound or a submicron filler such as talc, calcium carbonate or mica, which usually have been treated, for instance by grinding, to obtain particles of small, i.e. submicron, dimensions, *in situ* as stated above.

[00039] It is preferred that the filler (C) is layered silicate material, still more preferred, filler (C) is a clay-based compound. Clay-based compounds upon compounding of the polymer composition are dispersed in the polymer composition so that individual platelets in the layered structure are separated.

[00040] In a further preferred embodiment, the filler (C) is a clay-based layered inorganic, preferably silicate material or material mixture. Such useful clay materials include natural, synthetic and modified phyllosilicates. Natural clays include smectite clays, such as montmorillonite, hectorite, mica, vermiculate, bentonite. Synthetic clays include synthetic mica, synthetic saponite, synthetic hectorite. Modified clays include fluorinated montmorillonite, fluorinated mica.

[00041] Of course, the filler (C) may also contain components comprising a mixture of different fillers, such as mixtures of a clay-based filler and talc.

[00042] Layered silicates may be made organophylic before being dispersed in the polymer composition by chemical modification, such as by cation-exchange treatment using alkyl ammonium or phosphonium cation complexes. Such cation complexes intercalate between the clay layers.

[00043] Preferably, a smectite type clay is used, which comprises montmorillonite, beidellite, nontronite, saponite, as well as hectorite. The most preferred smectite type clay is montmorillonite.

[00044] Preferably, also talc is used as a filler (C).

[00045] The density affects most physical properties like stiffness impact strength and optical properties of the end products. Hence, and according to the present invention, the density of the polymer composition shall be of 945 kg/m³ or lower. More preferably, the density shall

range from 905 to 935 kg/m³, still more preferably from 910 to 930 kg/m³ and most preferably from 915 to 925 kg/m³.

[00046] The ranges and values given for the density in the whole invention apply for pure polymer compositions and do not include any additives, in particular no filler (C). The density is determined according to ISO 1183-1987.

[00047] Moreover, it is preferred that the polymer composition without any additive, preferably without filler (C) has a melt flow rate MFR₂ according to ISO 1133 at 190°C of 5 to 20 g/10 min, more preferably from 7 to 15 g/10 min.

[00048] Preferably, the polymer composition without any additive, preferably without filler (C) has a melt flow rate MFR₅ according to ISO 1133 at 190°C of 20 to 40 g/10 min, more preferably of 25 to 35 g/10 min.

[00049] Moreover, it is preferred that the melt flow ratio, which is a ratio of two melt flow rates measured for the same polymer under two different loads, falls within a specific range. The preferred specific range is 2.5 to 4.5, more preferably 2.7 to 4.0, for the melt flow ratio MFR₅/MFR₂.

[00050] A further characteristic of the molecular weight distribution (MWD) which is the relation between the number of molecules in a polymer and their individual chain length has to be considered. The width of the distribution is a number as a result of the ratio of the weight average molecular weight divided by the number average molecular weight (M_w/M_n). In the present invention, it is preferred that the polymer composition without any additive, preferably without filler (C), has a M_w/M_n of preferably 8 to 25 and more preferably from 10 to 20.

[00051] Additional additives, e.g. inorganic additives, known as excipients and extrusion aids in the field of coatings and films, are used.

[00052] For a better adhesion between the coating and the substrate, it is preferred that the polymer is oxidized. Consequently, it is preferred that the polymer composition contains anti-oxidants and process stabilizers less than 2,000 ppm, more preferably less than 1,000 ppm and most preferably not more than 700 ppm. The anti-oxidants thereby may be selected from those

known in the art like those containing hindered phenols, secondary aromatic amines, thio-ethers or other sulfur-containing compounds, phosphites and the like including their mixtures.

[00053] It has been found that the polymer composition as described above has a very low water-vapor transmission rate (WVTR). Additionally, the composition has a good adhesion to the substrate, in particular to aluminum, without any need to have an adhesion layer between the substrate and the coating. Further, the tendency of the coated article to curl is significantly reduced for the polymer composition compared to neat polymer. These advantageous effects could only be achieved as the miscibility between the polymer and the filler is much higher for a multimodal or bimodal polymer having a low molecular weight polymer fraction in comparison with a polymer having the same melt index and density.

[00054] In one preferable embodiment, the multimodal composition comprises as polymer (A), which is the low molecular weight fraction, a polyolefin (1), more preferably a low density polyethylene (LDPE) or linear low density polyethylene (LLDPE). The polyolefin (B), which is the high molecular weight fraction, is a low density polyethylene (LDPE) or a linear low density polyethylene (LLDPE). Preferably, this composition comprises a further polymer (A) which is a wax (2) as defined above. This composition can be produced in an *in situ* process or can be blended mechanically. Preferred properties for the polymer (A), in particular the polyolefin (1), the wax (2) and the polyolefin (B) are those as given above. In case this composition comprises two polymers (A), namely a polyolefin (1) and a wax (2), it is preferred that the amount of wax (2) in the total composition without filler (C) is 1 to 30 wt%, more preferably 1 to 20 wt% and most preferably 1 to 10 wt%. In turn, the composition comprises 70 to 99 wt%, more preferably 80 to 99 wt% and most preferably 90 to 99 wt% of LLDPE resulting from polymer (A) and polyolefin (B). In case the composition comprises LDPE, it is preferred that wax (2) is present in the amount of 1 to 30 wt% and LDPE resulting at least from polymer (B) and optionally from polymer (A) and is present in the amount of 70 to 99 wt% in the total composition without filler (C).

[00055] In another preferable embodiment, a polymer composition is produced in an *in situ* process, whereby the sequential step process by utilizing reactors coupled in series as described above is preferred. Preferably polymer (A) is produced in a loop reactor whereas

polyolefin (B) is produced in a gas phase reactor in the presence of polymer (A). Thereby, it is preferred that the multimodal polymer is at least a bimodal polymer. More preferably, polymer (A) and polyolefin (B) are both polyolefins. The polymer composition of this embodiment comprises 50 to 99 wt% of a linear low density polyethylene (LLDPE) having a multimodal, more preferably a bimodal molecular weight distribution (MWD) and more preferably 1 to 50 wt% of a filler (C), preferably a plate- or sheet-like filler such as mica or talc as described above.

[00056] In the following, when the description refers to LLDPE, it means that a multimodal polymer, preferably bimodal LLDPE is used, which comprises a low molecular weight (LMW) fraction, which is polymer (A) (polyolefin (1)), and a high molecular weight (HMW) fraction, which is polymer (B).

[00057] Preferably, the linear low density polyethylene (LLDPE) has a melt index MFR₂ from 1.0 to 20 g/10 min, more preferably from 2 to 15 g/10 min and most preferably from 3 to 10 g/10 min. It is preferred that the linear low density polyethylene (LLDPE) is at below 945 and ranges preferably from 905 to 935 kg/m³, more preferably from 910 to 930 kg/m³, most preferably from 915 to 925 kg/m³. If the melt index of the linear low density polyethylene (LLDPE) is lower than 1 g/10 min, a high throughput is not reached. On the other hand, if the melt index MFR₂ is higher than 20, the melt strength of the polyethylene suffers.

[00058] In addition, it is preferred that the linear low density polyethylene (LLDPE) has a melt flow index MFR₅ from 20 to 40 and preferably a melt flow ratio MFR₅/MFR₂ from 2.5 to 4.5, more preferably from 2.7 to 4.0. Furthermore, it is preferred that the linear low density polyethylene (LLDPE) has a weight average molecular weight (M_w) from 50,000 to 150,000 g/mol, more preferably from 60,000 to 100,000 g/mol and preferably a ratio of the weight average molecular weight to the number average molecular weight M_w/M_n of 8 to 25, more preferably of 10 to 20.

[00059] Moreover, the linear low density polyethylene (LLDPE) contains comonomers selected from the group consisting of C₃ alpha-olefin, C₄ alpha-olefin, C₅ alpha-olefm, C₆ alpha-olefin, C₇ alpha-olefin, C₈ alpha-olefin, C₉ alpha-olefin, C₁₀ alpha-olefin, C₁₁ alpha-olefin, C₁₂ alpha-olefin, C₁₃ alpha-olefin, C₁₄ alpha-olefin, C₁₅ alpha-olefin, C₁₆ alpha-olefin, C₁₇ alpha-olefin, C₁₈ alpha-olefin, C₁₉ alpha-olefin, C₂₀ alpha-olefin. Especially preferred are alpha-olefins

selected from the group consisting of propylene, 1-butene, 1-pentene, 4-methyl-1-pentene, 1-hexene, 1-heptene, 1-octene, 1-nonene, 1-decene, 6-methyl-1-heptene, 4-ethyl-1-hexene, 6-ethyl-1-octene and 7-methyl-1-octene. Still more preferred, alpha-olefins are selected from the group consisting of 1-butene, 4-methyl-1-pentene, 1-hexene and 1-octene.

[00060] As one requirement of the preferred embodiment is that the polymer composition is a linear low density polyethylene (LLDPE) the content of the comonomer units in the polymer is preferably 0.1 to 1.0 mol%, more preferably 0.15 to 0.5 mol%.

[00061] It is preferred that the linear low density polyethylene (LLDPE) without filler (C) comprises 40 to 60 wt%, more preferably 49 to 55 wt% polymer (A) and 60 to 40 wt%, and more preferably 51 to 45 wt% polyolefin (B).

[00062] As stated above, it is preferred that the linear low density polyethylene (LLDPE) comprises a LMW fraction, which forms the polymer (A). More preferably, the polymer (A), is a polyolefin (1), most preferably an ethylene copolymer containing alpha-olefins other than ethylene and listed above. Furthermore, it is preferred that the polymer (A) of the linear low density polyethylene (LLDPE) has a weight average molecular weight (M_w) of 10,000 to 60,000 g/mol, more preferably from 20,000 to 50,000 g/mol. It is further preferred that polymer (A) of the linear high density polyethylene (LLDPE), has a density of at least 905 to 935 kg/m³, more preferably of at least 910 to 930 kg/m³. In addition, it is preferred that polymer (A) of the linear low density polyethylene (LLDPE) has a melt flow rate MFR₂ from 1.0 to 20.0 g/10 min, more preferably from 2.0 to 15.0 g/10 min and most preferred from 3 to 10 g/10 min.

[00063] It is preferred that polyolefin (B) as the linear low density polyethylene (LLDPE) is an ethylene copolymer containing one or more alpha-olefins as listed above. Thereby, it is preferred that the amount of comonomer units in polyolefin (B) is from 2.0 to 15.0 mol%, more preferably from 3.0 to 10.0 mol%. In addition, it is preferred that the polyolefin (B) in the linear low density polyethylene (LLDPE) has a weight average molecular weight from 80,000 to 300,000 g/mol, more preferably from 100,000 to 200,000 g/mol.

[00064] The filler (C) and other additional components in the linear low density polyethylene (LLDPE) are identically used as listed and described above. It is in particular

preferred that additionally to the LLDPE, a wax (2), more preferably a polypropylene wax (2a) or an alkyl-ketene dimer (2b) as defined above is used as an additional polymer (A).

[00065] In case two polymers (A) are used, namely polyolefin (1) and wax (2), the amount of wax (2) is 1 to 30 wt%, more preferably 2 to 20 wt% and most preferably 1 to 10 wt% in the total composition without filler (C). In turn, the composition without filler (C) comprises 70 to 99 wt%, more preferably 80 to 88 wt% and most preferably 90 to 99 wt% LLDPE resulting from polymer (A) and polyolefin (B).

[00066] The further preferable embodiment of the present invention is a polymer composition whereby polymer (A) and polyolefin (B) are preferably mechanically blended. Thereby it is preferred that polymer (A) is a wax (2), more preferably a polypropylene wax (2a) or an alkyl-ketene dimer wax (2b).

[00067] In case of polymer (A), where a polypropylene wax (2a) is used, it is preferred that this wax (2a) has a weight average molecular weight (M_w) of 100 to 50,000, more preferably from 100 to 10,000, and most preferably from 5,000 to 6,000. In addition, it is preferred that the z-average molecular weight of the polypropylene wax (2a) ranges from 100 to 60,000 g/mol, and more preferably from 100 to 10,000 g/mol. It is preferred that the polypropylene wax (2a) has a number average molecular weight (M_n) of 100 to 2,000 g/mol, more preferably 500 to 3,000 g/mol. The melting temperature in DSC-analysis of the polypropylene wax (2a) is preferably of 95 to 130°C, more preferably 105 to 115°C.

[00068] Preferably, the polypropylene wax (2a) is mechanically blended with an ethylene polymer as a polyolefin (B) having an MFR₂ of 6.5 to 8.5 g/10 min, more preferably from 7 to 8 g/10 min and a density of 900 to 940 kg/m³, more preferably from 915 to 925 kg/m³. It is in particular preferred that polyolefin (B) is a low density polyethylene (LDPE), or a linear low density polyethylene (LLDPE) as described above.

[00069] The mechanically blended polymer including a talc as filler (C) and CaO as a water-absorbent component has preferably a density ranging from 1,000 kg/m³ to 1,300 kg/m³, more preferably of 1,150 to 1,200 kg/m³ and a melt flow rate MFR₂ of preferably 8 to 9.5 g/10 min, and more preferably of 8.5 to 9.0 g/10 min.

[00070] The other preferred alternative of a mechanical blend of wax (2) with polyolefin (B) is to use an alkyl-ketene dimer (2b) as wax (2). Preferably, this alkyl-ketene dimer (2b) has a weight average molecular weight (M_w) of 300 to 400 g/mol, more preferably from 320 to 350 g/mol. Preferably, the z-average molecular weight of the alkyl-ketene dimer (2b) is from 300 to 400 g/mol, more preferably from 360 to 390 g/mol. It is preferred that the alkyl-ketene dimer (2b) has a number average molecular weight (M_n) of 200 to 450 g/mol, more preferably from 280 to 300 g/mol. In addition, it is preferred that the alkyl-ketene dimer (2b) has a melting temperature DSC-analysis of 55 to 70°C, more preferably from 60 to 65°C.

[00071] For polyolefin (B), the same ethylene polymer is used as defined under the mechanical blend comprising a polypropylene wax (2a).

[00072] The density of the mechanically blended polymer composition comprising an alkyl-ketene dimer (2b) as defined above, an ethylene polymer (B) as defined above, a filler (C) and a water-absorbent component has preferably a density of 1,050 to 1,300 kg/m³ and more preferably from 1,150 to 1,250 kg/m³. The melt flow rate MFR₂ of this polymer composition is preferably from 12.5 g/10 min to 14.5 g/10 min and more preferably from 13 to 14 g/10 min. It is preferred that for this embodiment for filler (C) talc is employed and for the water absorbent compound CaO.

[00073] Furthermore, the present invention comprises a process for producing the multimodal composition as defined above.

[00074] A multimodal or at least bimodal, e.g. bimodal or trimodal, polymer may be produced by blending two or more monomodal polymers having differently centered maxima in their molecular weight distributions. The blending may be effected mechanically, e.g. analogously to the mechanical blending principles known in the art. Alternatively, the multimodal or at least bimodal, e.g. bimodal or trimodal, polymer composition may be produced by polymerization using conditions which create a multimodal or at least bimodal, e.g. bimodal or trimodal, polymer composition, i.e. using a catalyst system for mixtures with two or more different catalytic sides, using two or more stage polymerization process with different process conditions in the different stages (i.e. different temperatures, pressures, polymerization media, hydrogen partial pressures, etc.). With the polymer as produced in such a sequential step

process, i.e. by utilizing reactors coupled in series, and using different conditions in each reactor, the different polymer fractions produced in the different reactors will each have their own molecular weight distribution which may differ considerably from one another. The molecular weight distribution curve of the resulting final polymer can be regarded as superimposing of the molecular weight distribution curves of the polymer fractions which will accordingly show two or more distinct maxima, or at least the distinctively broadened maxima compared with the curves for individual fractions.

[00075] A polymer showing such a molecular weight distribution curve is called multimodal, trimodal or bimodal.

[00076] Multimodal polymers can be produced according to several processes, which are described, e.g. in WO 92/12182 and WO 97/22633.

[00077] A multimodal polymer is preferably produced in a multi-stage process in a multi-stage reaction sequence, such as described in WO 92/12182. The contents of this document are included herein by reference.

[00078] It is known to produce multimodal or at least bimodal, e.g. bimodal or trimodal, polymers, preferably multimodal or bimodal olefin-polymers, such as multimodal or bimodal polyethylenes in two or more reactors connected in series whereby the compounds (A) and (B) can be produced in any order.

[00079] According to the present invention, the main polymerization stages are preferably carried out as a combination of a slurry gas/gas-phase polymerization. The slurry polymerization is preferably performed in a so-called loop-reactor.

[00080] Optionally, and of more advantage, the main polymerization stages may be preceded by a pre-polymerization in which case up to 20 wt%, preferably 1-10 wt%, more preferably 1-5 wt% of the total amount of polymer composition is produced. At the pre-polymerization point, all of the catalyst is preferably charged into a loop-reactor and a polymerization is performed as a slurry polymerization. Such a polymerization leads to less fine particles being produced in the following reactors and to a more homogeneous product being obtained in the end. Such a pre-polymerization is for instance described in WO 96/18662.

[00081] Generally, the technique results in a multimodal or at least bimodal, e.g. bimodal or trimodal, polymer composition thereby a Ziegler-Natta or metallocene catalyst in several successive polymerization reactors is used. For example in the production of a bimodal high-density polyethylene composition, a first ethylene polymer is produced in the first reactor under certain conditions with respect to the hydrogen-gas concentration, temperature, pressure and so forth. After the polymerization the reactor-polymer including the catalyst is separated from the reaction mixture and transferred to a second reactor where further polymerization takes place under other conditions.

[00082] The components (A) and (B) can be produced with any suitable catalyst system, preferably a coordination catalyst, such as a Ziegler-Natta catalyst system, preferably a coordination catalyst, such as a Ziegler-Natta catalyst of a transition metal of a group 3-10 of the periodic table (TUPAC), a metallocene, non-metallocene, in a manner known in the art. One example of a preferred Ziegler-Natta catalyst comprises Ti, Mg and Al, such as described in document EP 0 688 794 B1, which is included herewith by reference. It is a high-activity procatalyst comprising a particular inorganic support, a curing compound deposited on the support, wherein the curing compound is the same as or different from the titanium compound, whereby the inorganic support is contacted with an alkyl metal chloride which is soluble in a non-polar hydrocarbon solvent, and has the formula $(R_nMeCl_{3-n})_m$, wherein R is a C_1 to C_{20} alkyl group, Me is a metal of Group III(13) of the periodic table, $n = 1$ or 2 and $m = 1$ or 2 , to give a first reaction product, and the first reaction product is contacted with a compound containing hydrocarbyl and hydrocarbyl oxide linked to magnesium which is soluble in non-polar hydrocarbon solvents, to give a second reaction product, and the second reaction product is contacted with a titanium compound which contains chlorine, having the formula $Cl_xTi(OR^{IV})_{4-x}$, wherein R^{IV} is a C_2 to C_{20} hydrocarbyl group and $x = 3$ or 4 , to give the procatalyst. Preferred supports are inorganic oxides, more preferably silicon dioxide or silica. Most preferably silica having an average particle size of $20\ \mu m$ is used. Even more preferred tri-ethyl aluminium as a co-catalyst is used. Alternatively, a metallocene of group 4 metal can be used.

[00083] Preferably, polymer (A), the low molecular weight (LMW) polymer, is produced with addition or no addition of comonomer in a first reactor, and also the polyolefin (B), the high

molecular weight (HMW) polymer, is produced with addition or no addition, more preferably with addition, of comonomer in the second reactor.

[00084] The resulting end product consists of an intimate mixture of polymers from the two reactors, the different molecular weight distribution occurs of these polymers together forming a molecular weight distribution curve having a broad maximum or two maxima, i.e. the end product is a multimodal or bimodal polymer mixture. Since multimodal and, in particular, bimodal polymers, preferably ethylene polymers and the production thereof belong to the prior art, no detailed description is called for here, but reference is made to the above-mentioned document WO 92/12182. It will be noted that the order of the reaction stages may be reversed.

[00085] Preferably, as stated above, the multimodal polymer composition according to the invention is a bimodal or trimodal polymer composition. It is also preferred that this bimodal or trimodal polymer composition has been produced by polymerization as described above under different polymerization conditions in two or more polymerization reactors connected in series.

[00086] Furthermore, it is preferred that for the multimodal composition according to this invention a process is used as defined above whereby polymer (A) and polyolefin (B) are produced together in a multi-stage process comprising a loop reactor and a gas-phase reactor, wherein polymer (A) is generated in at least one loop reactor and the polyolefin (B) is generated in a gas-phase reactor in the presence of the reaction product (A) of the loop reactor, and filler (C) and the composition comprising polymer (A) and polyolefin (B) are blended together and compounded.

[00087] In particular, a multi-stage process is used as described above. Especially, it is preferred that a loop reactor is operated at 75 to 100°C, more preferably in the range of 85 to 100°C and most preferably in the range of 90 to 98°C. Thereby, the pressure is preferably 58 to 68 bar, more preferably 60 to 65 bar.

[00088] Preferably, polymer (A) is prepolymerized in a first loop reactor and then continuously removed to a second loop reactor where the polymer (A) is further polymerized. It is preferred that the temperature in the second loop reactor is 90 to 98°C, more preferably about 95°C. Thereby, the pressure is preferably 58 to 68 bar, more preferably about 60 bar.

[00089] In addition, it is preferred that in the second loop reactor, the ethylene concentration is 4 to 10 mol%, more preferably 5 to 8 mol% and most preferably about 6.7 mol%.

[00090] The hydrogen to ethylene mol-ratio highly depends on the catalyst used. It must be adjusted to render the desired melt flow rate MFR of the polymer withdrawn from the loop reactor. For the preferred catalyst as described it is preferred that the ratio of hydrogen to ethylene is 100 to 800 mol/kmol and more preferably 300 to 700 mol/kmol, still more preferably 400 to 650 mol/kmol and most preferred about 550 mol/kmol.

[00091] The polymer slurry is then preferably removed from the loop reactor by using settling tanks and is then preferably introduced into a flash vessel operating preferably at about 3 bar pressure, where the polymer is separated from most of the fluid phase. The polymer is then preferably transferred into a gas-phase reactor operating preferably at 75 to 95°C, more preferably 80 to 90°C and most preferably about 85°C, and at preferably 10 to 50 bar, more preferably 15 to 25 bar and most preferably about 20 bar.

[00092] Additionally, ethylene comonomers were used and hydrogen as well as nitrogen as an inert gas are preferably introduced into the reactor so that the fractional ethylene in the fluidization gas is preferably 1 to 10 mol%, more preferably 1 to 5 mol% and most preferably about 2.5 mol% and the ratio of hydrogen to ethylene is preferably 100 to 400 mol/kmol, more preferably 150 to 300 mol/kmol and most preferably about 210 mol/kmol.

[00093] The comonomer to ethylene ratio has influence on the desired density of the bimodal polymer. Hence, it is preferred that the ratio of comonomer to ethylene is 20 to 150 mol/kmol, more preferably 50 to 100 mol/kmol and most preferably about 80 mol/kmol. Preferably, after the polymer is withdrawn from the gas-phase reactor and then mixed with further additives as anti-oxidants and/or process stabilizers by blending.

[00094] The polymer mix of polymer (A) and polyolefin (B) is then blended with filler (C) and with any suitable method known in the art. These methods include compounding in a twin-screw extruder, like a counter-rotating twin-screw extruder or a co-rotating twin-screw extruder and compounding in a single-screw extruder.

[00095] In addition, the present invention comprises a new multi-layer material comprising at least a substrate as a first layer (I) and a multimodal polymer composition as described above as at least one further layer (II). Preferably, the multi-layer material consists of a substrate as a first layer (I) and a multimodal polymer composition as described above as at least one further layer (II).

[00096] It is further preferred that the multi-layer material is a two-layer or three-layer material consisting of a substrate as a first layer and of a polymer composition for the second and third layer, whereby preferably at least the second layer is a polymer composition as defined above. The layers can of course be in any order. Optionally, this multi-layer material comprises adhesion promoters as tetra-iso-propyl titanate, tetra-stearyl titanate, tetrakis(2-ethylhexyl) titanate, poly(dibutyltitanate).

[00097] Preferably, the substrate is selected from the group consisting of paper, paper-board, aluminum film and plastic film.

[00098] Preferably, the multi-layer material comprises as a further layer (III) a low density polyethylene (LDPE). Thereby, it is preferred that the low density polyethylene has a density of 900 to 950 kg/m³, more preferably from 915 to 925 kg/m³. In addition, it is preferred that the melt flow rate MFR₂ of the low density polyethylene (LDPE) is of 2.0 to 20.0 g/10 min, more preferably from 3.0 to 10.0 g/10 min.

[00099] Preferably, the coating weight of layer (II) comprising the polymer composition according to the present invention ranges from 5 to 60 g/m² and more preferably from 10 to 45 g/m². Additionally, it is preferred that the layer (III) comprising a low density polyethylene (LDPE) as described above has a coating weight of 0 to 25, more preferably from 3 to 18 g/m².

[000100] The present invention also comprises a film, preferably a cast film, comprising the multimodal polymer composition as described above, more preferably, the film consists of the multimodal polymer composition of the present invention.

[000101] Furthermore, the present invention provides a process for producing a multi-layer material comprising the inventive polymer composition as described above. Thereby, it is preferred that the multimodal polymer composition as described above is applied on a substrate

by a film-coating line comprising an unwind, a wind, a chill roll, and a coating die. Preferably, the speed of the coating line ranges from 50 to 5,000 m/min, more preferably from 100 to 1,500 m/min. The coating may be done as any coating line known in the art. It is preferred to employ a coating line with at least two extruders to make it possible to produce multi-layered coatings with different polymers. It is also possible to have arrangements to treat the polymer melt exiting the die to improve adhesion, e.g. by ozone treatment, corona treatment or flame treatment.

[000102] In addition, the present invention comprises the use of the multimodal polymer composition as defined above for extrusion coating, in particular for extrusion coating producing a multi-layer material as described above.

[000103] Furthermore, the present invention relates to the use of the multimodal polymer composition for films, preferably cast films.

DETAILED DESCRIPTION OF THE INVENTION

[000104] In the following the present invention is demonstrated by means of examples.

Examples

Measurements

[000105] WVTR: Water vapor transmission rate was measured at 90 % relative humidity and 38 °C temperature according to the method ASTM E96.

[000106] Basis weight or coating weight: Basis weight (or coating weight) was determined as follows: Five samples were cut off from the extrusion coated paper parallel in the transverse direction of the line. The size of the samples was 10 cm x 10 cm. The samples were dried in an oven at 105 °C for one hour. The samples were then weighed and the coating weight was calculated as the difference between the basis weight of the coated structure and the basis weight of the substrate. The result was given as a weight of the plastic per square meter.

[000107] Molecular weight averages and molecular weight distribution: Molecular weight averages and molecular weight distribution were determined by ISO 16014, part 2 universal

calibration (narrow MWD polystyrene standards (universal alibration) and a set of 2 x mixed bed + 1 x 10⁷ A Tosohas (JP) columns were used).

[000108] Density: Density was determined according to ISO 1183-1987.

[000109] Melt flow rate or melt index: Melt flow rate (also referred to as melt index) was determined according to ISO 1133, at 190 °C. The load used in the measurement is indicated as a subscript, i.e. MFR₂ denotes the MFR measured under 2.16 kg load.

[000110] Flow rate ratio: Flow rate ratio is a ratio of two melt flow rates measured for the same polymer under two different loads. The loads are indicated as a subscript, i.e., FRR_{5/2} denotes the ratio of MFR₅ to MFR₂.

[000111] Curling: Curling was determined by cutting a circular sample having an area of 100 cm² within two hours after the coating. The sample is then allowed freely to curl at the table for two minutes. The curl is then measured as the difference (in mm) from the table to the curled sheet.

Example 1

[000112] A dry blend of pellets was made of 650 kg of the low density polyethylene CA8200 of 300 kg of a talc filler Finntalc MO5SL, manufactured and sold by Mondo Minerals and 50 kg of Clariant PP6100 PP wax. This dry blend was then compounded and pelletized by using the above-mentioned ZSK70 extruder. The melt temperature during the extrusion was 200°C. The composition was then dried at 60°C for 6 hours to remove the moisture. CA8200 is a low density polyethylene designed for extrusion coating, produced and marketed by Borealis. It is produced by free radical polymerization in a high pressure autoclave process. It has an MFR₂ of 7.5 g/10 min and a density of 920 kg/m³. Clariant PP6100 is a low molecular weight propylene polymer having a number average molecular weight of 2,090 g/mol, weight average molecular weight 5,370 g/mol, z-average molecular weight 10,900 g/mol and melting temperature in DSC analysis 109°C. The composition had a density of 1,195.7 kg/m³ and MFR₂ of 6.1 g/10 min.

Comparative Example 1

[000113] The procedure of Example 1 was repeated, except that the amount of CA8200 was 700 kg and Clariant PP6100 was not used. Moreover, no drying at 60°C was done.

Table 1: Data for compositions containing polyolefin and talc used in cast films.

Example	Composition	MFR ₂ g/10 min	Density 920 kg/m ³
Example 1	LD/PP/talc	NA	NA
Comparative Example 1	LD/ - /talc	NA	NA

Example 2

[000114] The composition of Example 1 was used to make a cast film on Collin laboratory scale cast film line, having a single screw extruder with a screw diameter of 30 mm and length to diameter (L/D) ratio of 30. The line speed was about 10 m/s (from 8.9 to 10.3 m/s), the output about 5 kg/h (from 4.91 to 6.07 kg/h), the die temperature 250°C and melt temperature 245°C. The temperature of the chill roll was about 70°C (68 to 72°C). The data can be found in Table 2. The thickness of the film was 45 µm. The WVTR was 5.0 g/m²/24 h.

Example 3

[000115] The procedure of Example 2 was repeated, except that the thickness of the film was 98 µm. The WVTR was 2.3 g/m²/24 h.

Comparative Example 2

[000116] The procedure of Example 3 was repeated, except that the composition of Comparative Example 1 was used in place of the composition of Example 1. Data can be found in Table 2.

Table 2: Cast film data.

Example	Composition	Thickness μm	WVTR $\text{g/m}^2/24 \text{ h}$
Example 2	LD/ PP/ talc	45	5.0
Example 3	LD/ PP/ talc	98	2.3
Comparative Example 2	LD/ - /talc	102	2.7

ABSTRACT

The present invention relates to a polymer composition with good chemical properties and barrier properties being multimodal and comprising a polymer (A) having a weight average molecular weight of lower than 6,000 g/mol and a polyolefin (B) having a higher weight average molecular weight than polymer (A) and a filler (C), whereby a polymer composition without filler (C) has a density of 940 kg/m³ or lower.

EXTRUSION COATING POLYETHYLENE

FIELD OF THE INVENTION

[0001] The present invention relates to a polymer composition suitable for extrusion coating and films, preferably cast films having good chemical properties and barrier properties, in particular, a low water-vapor transmission rate (WVTR) and a low curling. Additionally, the present invention relates to the process for producing the inventive composition and its use. Moreover, the present invention is related to a multi-layer material comprising the polymer composition as well as to a process of said multi-layer material.

BACKGROUND OF THE INVENTION

[0002] One of the largest and most rapidly growing polyolefin-processing method is extrusion coating. The largest single volume of coated materials are different papers and paperboards, which are used for a variety of packaging applications. Other material frequently coated are polymer films, cellophane, ~~aluminum~~ aluminum foil, freezer wrap paper and fabrics of various kinds. One target for the improvement of coated articles is to reduce the water-vapor transmission rate (WVTR) as much as possible. A coated material with a low water-vapor transmission rate (WVTR) can for example protect the products wrapped therein much better. The demanded requirement applies, of course, not only to coated materials but also to cast films used for packaging or containers. In both cases, a low water-vapor transmission rate is required. Much effort has been undertaken to improve the water-vapor transmission rate of coated materials as well as for cast films. To date, several new polymer compositions have been developed and much effort has been undertaken to find appropriate fillers to improve the barrier properties significantly. Furthermore, different polymers have been designed as cyclo-olefin copolymers (COC) and liquid crystal polymers (LCP). However, these materials have the drawback of being expensive and having minor processability properties.

[0003] WO 00/71615 discloses for example the use of a bimodal high density polyethylene (HDPE) with a melt flow rate, MFR₂, of 5 g/10 min and a density of 957 kg/m³ for extrusion coating. No information is given how to improve the water-vapor transmission rate (WVTR).

[0004] WO 00/34580 describes release liner for pressure-sensitive adhesive labels. The release-liner contains a paper wrap, a filled polymer layer, and, on the opposite of the paper web, an extrudate, e.g. polyethylene, and on the top of the extrudate, a release film. The filled polymer layer can be polyethylene and the filler is an inert particulate, such as silica, mica, clay, talc and titanium oxide. The filler is present in 15 to 40 wt% of the composition.

[0005] US 4,978,572 describes a laminated film having three layers. The first layer comprises a thermoplastic resin and 0.3 to 30 wt% white inorganic particles. The second ~~one~~ layer comprises an ethylene copolymer, 0.5 to 90 wt% of a substance giving anti-block action and anti-oxidant. The third ~~one~~ layer comprises a metallized thermoplastic. The substance giving anti-block action of the second layer may be silica or talc. The laminated film is reported to have good mechanical strength and good barrier properties.

[0006] Even though the prior art offers already a variety of products having good water-vapor transmission rates (WVTR), there is still demand for a significant improvement of these properties. One significant disadvantage in polymer compositions comprising fillers reducing the water-vapor transmission rate (WVTR) is the low dispersion of the fillers incorporated in the polymer matrix. Conventional mechanical incorporation frequently results in poor dispersion as usual fillers form multi-layer aggregation caused by incompatibility with polymer matrix. One consequence of the described phenomenon is that the water-vapor transmiss-ion rate (WVTR) varies considerably in the layer leading to unsatisfying average values for the WVTR. Secondly, the low dispersion of the filler causes an easy upcurling of the polymer composition coated on the materials. Hence, a uniform dispersion of fillers incorporated in a polymer composition should improve the water-vapor transmission rate significantly, and, additionally, the curling properties of a coated material should be enhanced.

[0007] Hence, the object of the present invention is to improve the water-vapor transmission rate (WVTR).

[0008] The present invention is based on the finding that the object can be addressed by a polymer composition comprising a polymer having a low average molecular weight enabling an enhanced and uniform dispersion of fillers incorporated in the polymer composition.

SUMMARY OF THE INVENTION

[0009] The present invention therefore provides a multimodal polymer composition comprising a) at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60,000 g/mol; b) at least one polyolefin (B) having a higher weight average molecular weight (M_w) than polymer (A); and c) a filler (C) whereby the polymer composition without filler (C) has a density of 940 kg/m³ or lower.

[0010] It is preferred that the polymer composition consists of a) at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60,000 g/mol; b) at least one polyolefin (B) having a higher weight average molecular weight (M_w) than polymer (A); and c) filler (C) whereby the polymer composition without filler (C) has a density of 940 kg/m³ or lower.

[0011] Accordingly, the polymer composition according to this invention is multimodal with respect to the molecular weight distribution. "Multi-modal" or "multimodal distribution" describes a frequency distribution that has several relative maxima. In particular, the expression "modality of a polymer" refers to the form of its molecular weight distribution (MWD) curve, i.e. the appearance of the graph of the polymer weight fraction as a function of its molecular weight. The molecular weight distribution (MWD) of a polymer produced in a single polymerization stage using a single monomer mixture, a single polymerization catalyst and a single set of process conditions (i.e. temperature, pressure, etc.) shows a single maximum the breadth of which depends on catalyst choice, reactor choice, process conditions, etc., i.e. such a polymer is monomodal.

[0012] This inventive composition is characterized by a very low water-vapor transmission rate (WVTR) and also by low curling-values for extrusion-coated layers. These improved properties are reached by a much better dispersion of the filler (C) in the polymer mixture of polymer (A) and polyolefin (B) compared with an unimodal polymer having the same melt index and density for both extrusion-coated layers and cast films.

[0013] Hence, the polymer composition according to this invention is a multimodal **polymer** including bimodal polymer composition consisting of at least two different polymers

having two different molecular weight distribution curves and are blended mechanically or *in situ* during the preparation thereof. Preferably the polymer composition is at least a bimodal mechanical or *in-situ* blend of a polyolefin (1) (as polymer (A)) and polymer (B). In case such a bimodal blend comprises further a wax (2) as an additional polymer (A), then the final polymer composition may also be trimodal.

[00014] The molecular weight distribution (MWD) is the relation between the numbers or molecules in a polymer and their individual chain length. The molecular weight distribution (MWD) is often given as a number, which normally means weight average molecular weight (M_w) divided by number average molecular weight (M_n).

[00015] The weight average molecular weight (M_w) is the first moment of a plot of the weight of polymers in each molecular weight range against molecular weight. In turn, the number average molecular weight (M_n) is an average molecular weight of a polymer expressed as the first moment of a plot of the number of molecules in each molecular weight range against the molecular weight. In effect, this is the total molecular weight of all molecules divided by the number of molecules.

[00016] The number average molecular weight (M_n) and the weight average molecular weight (M_w) as well as the molecular weight distribution (MWD) are determined according to ISO 16014.

[00017] The weight average molecular weight (M_w) is a parameter for the length of the molecules in average. Low M_w -values indicate that the chain length of the molecules is rather short in average. It has been found out that a polymer mixture comprising a polymer (A) with M_w -values of lower than 60,000 g/mol contributes inter alia to better barrier properties and better dispersion of the filler (C). Such better dispersion improves the water-vapor transmission rate (WVTR) as well as the curling resistance positively.

[00018] Hence, as a further requirement of the present invention, the multimodal polymer composition must comprise at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60,000 g/mol. It is in particular preferred that at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60,000 g/mol is at least one

polyolefin (1) having a weight average molecular weight (M_w) of 10,000 to 60,000 g/mol, more preferably of 20,000 to 50,000 g/mol and/or at least one wax (2) having a weight average molecular weight (M_w) of less than 10,000 g/mol, more preferably in the range of 500 to 10,000 g/mol.

[00019] Moreover, it is preferred that the polyolefin (1) is a polyethylene or polypropylene, more preferably a polyethylene. The polyolefin (1) can be a homopolymer or copolymer. It is preferred that the polyolefin (1) is a homopolymer or copolymer of propylene or ethylene, more preferred the polyolefin (1) is a homopolymer or copolymer of ethylene. Most preferably the polyolefin (1) is a low density polyethylene (LDPE), a linear low density polyethylene (LLDPE), or a linear medium density polyethylene (LMDPE). LDPE, LLDPE and LMDPE are equally suitable alternatives for polyolefin (1), e.g. where a LLDPE or a LMDPE is applicable also a LDPE can be used and vice versa.

[00020] In case polymer (A) is a wax (2), it is preferred that it is selected from one or more of ~~2(a)~~ a polypropylene wax (2a) having a weight average molecular weight (M_w) of less than 10,000 g/mol, more preferably in the range of 500 to 10,000 g/mol, still more preferably in the range of 1,000 to 9,000 g/mol, yet more preferably in the range of 2,000 to 8,000 g/mol and most preferably in the range of 4,000 to 8,000 g/mol or a polyethylene wax having a weight average molecular weight (M_w) of less than 10,000 g/mol, more preferably in the range of 500 to 10,000 g/mol, still more preferably in the range of 1,000 to 9,000 g/mol, yet more preferably in the range of 2000 to 8000 g/mol and most preferably in the range of 4,000 to 8,000 g/mol, and ~~2(b)~~ an alkyl ketene dimer wax having weight average molecular weight (M_w) of less than 10,000 g/mol more preferably lower than 5000 g/mol yet more preferably lower than 1,000 g/mol. In turn the alkyl ketene dimer wax has preferably weight average molecular weight (M_w) of at least 100 g/mol. Most preferred the alkyl ketene dimer wax (2b) has weight average molecular weight (M_w) in the range of 250 to 1,000 g/mol.

[00021] The terms “at least one polymer (A)”, “at least one polyolefin (1)”, or “at least one wax (2)” shall indicate that more than one polymer (A), polyolefin (1) or wax (2) can be present in the multimodal polymer composition. It is preferred that three, two or one different polymers (A) as defined above are used in a multimodal polymer composition. Still more

preferred is that wax (2), preferably a polypropylene wax (2a) or an alkyl ketene dimer wax (2b) as defined above is used as a component (A) only. In case the component (A) comprises a polyolefin (1) as defined above, it is preferred that a wax (2) is present in the multimodal polymer composition as a further polymer (A). In such cases the multimodal composition is preferably trimodal comprising polyolefin (1), wax (2) and polyolefin (B) having different centered maxima in their molecular weight distribution, e.g. having different weight average molecular weights (M_w). The use of the wax (2) has the benefit that the amorphous region of the polymer matrix, which may be a mix of polyolefin (1) and polyolefin (B), is filled up and improves thereby the barrier properties.

[00022] It is preferred that not only the final polymer composition has a specific density of 940 kg/m^3 or lower but also the polymer (A) shall have a density of lower than 945 kg/m^3 . It is preferred that polyolefin (1) when used as polymer (A) has a density lower than 945 kg/m^3 , more preferably is in a range of 905 to 935 kg/m^3 , still more preferably in the range of 910 to 930 kg/m^3 and most preferred in the range of 915 to 925 kg/m^3 . Preferably, the polyolefin (1) is a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE). In turn, also a low density polyethylene (LDPE) produced in a high pressure process by free radical polymerization is applicable as a polyolefin (1). The linear low density polyethylene (LLDPE) or the linear medium density polyethylene (LMDPE) is produced by a process as described for the polyolefin (B) below.

[00023] The molecular weight distribution (MWD) of the polymer composition is further characterized by the way of its melt flow rate (MFR) according to ISO 1133 at 190°C . The melt flow rate (MFR) mainly depends on the average molecular weight. The reason for this is that long molecules give the material a lower flow tendency than short molecules.

[00024] An increase in molecular weight means a decrease in the MFR-value. The melt flow rate (MFR) is measured in g/10 min of the polymer discharged under specific temperature and pressure conditions and is the measure of a viscosity of the polymer which in turn for each type of polymer is mainly influenced by its molecular weight distribution, but also by its degree of branching. The melt flow rate measured under a load of 2.16 kg (ISO 1133) is denoted as MFR2. In turn, the melt flow rate measured with 5 kg load (ISO 1133) is denoted as MFR5.

[00025] In case polymer (A) is a polyolefin (1), it is preferred that MFR₂ is in the range of 1.0 to 20.0 g/10 min and more preferably in the range of 2.0 to 15.0 g/10 min and for example in the range of 3.0 to 10.0 g/10 min. According to one embodiment, the polyolefin (1) is a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE) with MFR₂ as given above. In turn, polyolefin (1) can also be a low density polyethylene (LDPE) having a density in the range as stated in this paragraph. The low density polyethylene (LDPE) is produced in a high-pressure process by free radical polymerization. In turn, the linear low density polyethylene (LLDPE) or the linear medium density polyethylene (LMDPE) is produced as described for polyolefin (B).

[00026] In case polymer (A) is an ethylene homopolymer, it is preferred that the ethylene homopolymer contains less than 0.2 mol%, more preferably less than 0.1 mol% and most preferably less than 0.05 mol% units derived from alpha-olefins other than ethylene. It is in particular preferred that the polymer (A) is an ethylene co-polymer, more preferably having a weight average molecular weight from 10,000 to 60,000 g/mol, still more preferably from 20,000 to 50,000 g/mol. Yet more preferably the polymer (A) is an ethylene copolymer having a density of 905 to 935 kg/m³, more preferably of 910 to 930 kg/m³, most preferably of 915 to 925 kg/m³ and having a melt index MFR₂ from 1.0 to 20.0 g/10 min, more preferably from 2.0 to 15.0 g/10 min, most preferably of 3.0 to 10.0 g/10 min. Preferably, the ethylene copolymer comprises, more preferably consists of, comonomer units as defined below for the LLDPE. It is in particular preferred that the ethylene copolymer fulfills all properties as stated above simultaneously.

[00027] In case polymer (A) is a wax (2a), namely a polypropylene wax or a polyethylene wax, it is preferred that the wax (2a) has a weight average molecular weight (M_w) in the range of 500 to 10,000 g/mol, more preferably in the range of 1,000 to 9,000 g/mol, still more preferably in the range of 2,000 to 8,000 g/mol and most preferably in the range of 4,000 to 8,000 g/mol. Further preferred ranges for the weight average molecular weight (M_w) of the wax (2a), in particular the polypropylene or polyethylene wax, is in the range of 4,000 to 7,000 g/mol, still more preferably in the range of 5,000 to 6,000 g/mol and most preferably in the range of 5,300 to 5,400 g/mol. Additionally, it is preferred that the wax (2a), in particular the polypropylene wax or polyethylene wax, has a z-average molecular weight of 9,100 to 40,000 g/mol, more

preferably from 500 to 20,000 g/mol and most preferably from 10,000 to 12,000 g/mol. It is additionally preferred that the wax (2a), in particular the polypropylene wax or the polyethylene wax, has a number average molecular weight (M_n) of 100 to 20,000 g/mol, more preferably of 500 to 3,000 g/mol.

[00028] Moreover, it is preferred that wax (2a), in particular polypropylene wax or polyethylene wax, has a specific molecular weight distribution (MWD) which is the relation between the number of molecules in the polymer and their individual chain length. The molecular weight distribution is given as a number which means weight average molecular weight divided by number average molecular weight (M_w/M_n). It is preferred that the wax (2a), in particular the polypropylene wax or the polyethylene wax, has an MWD in the range of 1 to 5, more preferably in the range of 1.5 to 4.

[00029] In addition, it is preferred that the wax (2a), in particular the polypropylene wax or the polyethylene wax, has a melting temperature in DSC-analysis of below 150°C, more preferably below 140°C, still more preferably in the range of 95 to 130°C, most preferably in a range of 105 to 115°C.

[00030] In case a wax (2b), namely an alkyl-ketene dimer, is employed as polymer (A), it is preferred that the weight average molecular weight (M_w) of the wax (2b) is higher than 100 g/mol. In turn, it is preferred that the weight average molecular weight of the wax (2b) is lower than 10,000 g/mol, more preferably lower than 5,000 g/mol, still more preferably lower than 1,000 g/mol. Preferred ranges for the weight average molecular weight (M_w) of the wax (2b) is 100 to 10,000 g/mol, more preferably 250 to 1,000 g/mol. Additionally, it is preferred that the wax (2b) has a number average molecular weight (M_n) of 100 to 20,000 g/mol, more preferably in the range of 100 to 800 g/mol. In addition, it is preferred that wax (2b) has a melting temperature in DSC-analysis below 140°C, more preferably below 100°C. A preferred range for the melting temperature in DSC-analysis is 50 to 90°C, more preferably 50 to 70°C.

[00031] As a further requirement, according to the present invention, the polyolefin (B) shall have a higher weight average molecular weight (M_w) than polymer (A). It is preferred that the polyolefin (B) has a weight average molecular weight (M_w) of higher than 80,000 g/mol, more preferably higher than 100,000 g/mol. The upper limit for the weight average molecular

weight for polyolefin (B) shall preferably not be higher than 300,000 g/mol, more preferably not higher than 200,000 g/mol. The preferred range for the weight average molecular weight for polyolefin (B) is 80,000 to 300,000 g/mol, more preferably from 100,000 to 200,000 g/mol. Preferably, polyolefin (B) is a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE), which has been preferably produced in a low medium pressure process in the presence of a polymerization catalyst (i.e. a Ziegler-Natta catalyst or a metallocene catalyst). A linear low density polyethylene (LLDPE) and a linear medium density polyethylene (LMDPE) has a density lower than 945 kg/m³, more preferably in the range of 905 to 935 kg/m³, still more preferably in the range of 910 to 930 kg/m³ and most preferred in the range of 915 to 925 kg/m³. However, also a low density polyethylene (LDPE) is also applicable for the polyolefin (B). A low density polyethylene (LDPE) has the same density ranges as the LLDPE or the LMDPE as stated in this paragraph and is a product from a high pressure polymerization process characterized by a highly branched chain structure. LDPE, LLDPE and LMDPE are equally suitable alternatives for polyolefin (B), e.g. where a LLDPE or a LMDPE is applicable also a LDPE can be used and vice versa.

[00032] According to this invention, more than one polyolefin (B) can be used. Accordingly, the invention also comprises the possibility of any mixture of a linear low density polyethylene (LLDPE), a linear medium density polyethylene (LMDPE) and a low density polyethylene (LDPE).

[00033] The MFR₂ of the polyolefin (B) is preferably in the range of 1.0 to 20.0 g/10 min, more preferably in the range of 2.0 to 15.0 g/10 min, in the range of e.g. 3.0 to 10.0 g/10 min. It is in particular preferred that the linear low density polyethylene (LLDPE) and the linear medium density polyethylene (LMDPE) have such melt flow characteristics. In turn, also the low density polyethylene (LDPE) suitable as a polyolefin (B) may have the melt flow characteristics as given in this paragraph.

[00034] It is preferred that the polyolefin (B) is a polyethylene. In case the polyolefin (B) is a polyethylene, it may be an ethylene homopolymer or an ethylene copolymer. In case for the polyolefin (B) an ethylene homopolymer is employed, then preferably an ethylene homopolymer is used as defined for polymer (A). In case an ethylene copolymer is employed

for polyolefin (B), then preferably an ethylene copolymer is used as defined below. It is in particular preferred that polyolefin (B) is a low density polyethylene (LDPE), a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE).

[00035] According to one embodiment, the polymer composition according to this invention is a linear low density polyethylene (LLDPE) comprising polyolefin (1) (polymer (A)) as a low molecular weight fraction of LLDPE and polyolefin (B) as a high molecular weight fraction of LLDPE. This linear low density polyethylene (LLDPE) may be a mechanical blend, preferably an in-situ blend produced in a ~~multistage~~ **multi-stage** process. Preferably said composition comprises wax (2) as a further polymer (A).

[00036] It is preferred that the polymer composition as defined above comprises 1 to 50 wt% of polymer (A), 40 to 90 wt% of polyolefin (B) and 1 to 50 wt% of filler (C), more preferably of 5 to 40 wt%, and most preferably of 10 to 35 wt%. In case the polymer composition is produced in an *in situ* polymerization process, e.g. a sequential step process by utilizing reactors coupled in series and described as above, it is preferred that the polymer (A) may range from 40 to 60 wt%, more preferably 49 to 55 wt% in the polymer mix without filler (C). In turn, it is preferred that in such a polymer mix, the polyolefin (B) ranges from 60 to 40 wt%, more preferably from 51 to 45 wt%. Preferably, the total polymer composition comprises 50 to 99 wt% of said polymer mix and of 1 to 50 wt% filler (C), more preferably of 5 to 40 wt%, and most preferably of 10 to 35 wt%.

[00037] In case polymer (A) and polyolefin (B) are blended mechanically, it is preferred that polymer (A) ranges from 1 to 30 wt% and, more preferably, from 1 to 20 wt% in the total polymer composition. These ranges apply in particular in case for polymer (A) a wax (2) is used only.

[00038] The last requirement according to the present invention is that the multimodal polymer composition additionally comprises a filler (C). Any filler having a positive influence on the water-vapor transmission rate (WVTR) can be used. Preferably, the filler shall be lamellar, such as clay, mica or talc. More preferably, the filler shall be finely divided. The finely divided filler consists of about 95 wt% of particles having particle sizes of less than 10 μm , and about 20-30 wt% of particles having a particle size of less than 1 μm . In the present

invention all layer materials may be used as long as they have the ability to disperse in the polymer composition. The filler may either be a clay-based compound or a submicron filler such as talc, calcium carbonate or mica, which usually have been treated, for instance by grinding, to obtain particles of small, i.e. submicron, dimensions, *in situ* as stated above.

[00039] It is preferred that the filler (C) is layered silicate material, still more preferred, filler (C) is a clay-based compound. Clay-based compounds upon compounding of the polymer composition are dispersed in the polymer composition so that individual platelets in the layered structure are separated.

[00040] In a further preferred embodiment, the filler (C) is a clay-based layered inorganic, preferably silicate material or material mixture. Such useful clay materials include natural, synthetic and modified phyllosilicates. Natural clays include smectite clays, such as montmorillonite, hectorite, mica, vermiculate, bentonite. Synthetic clays include synthetic mica, synthetic saponite, synthetic hectorite. Modified clays include fluorinated montmorillonite, fluorinated mica.

[00041] Of course, the filler (C) may also contain components comprising a mixture of different fillers, such as mixtures of a clay-based filler and talc.

[00042] Layered silicates may be made organophylic before being dispersed in the polymer composition by chemical modification, such as by cation-exchange treatment using alkyl ammonium or phosphonium cation complexes. Such cation complexes intercalate between the clay layers.

[00043] Preferably, a smectite type clay is used, which comprises montmorillonite, beidellite, nontronite, saponite, as well as hectorite. The most preferred smectite type clay is montmorillonite.

[00044] Preferably, also talc is used as a filler (C).

[00045] The density affects most physical properties like stiffness impact strength and optical properties of the end products. Hence, and according to the present invention, the density of the polymer composition shall be of 945 kg/m³ or lower. More preferably, the density shall

range from 905 to 935 kg/m³, still more preferably from 910 to 930 kg/m³ and most preferably from 915 to 925 kg/m³.

[00046] The ranges and values given for the density in the whole invention apply for pure polymer compositions and do not include any additives, in particular no filler (C). The density is determined according to ISO 1183-1987.

[00047] Moreover, it is preferred that the polymer composition without any additive, preferably without filler (C) has a melt flow rate MFR₂ according to ISO 1133 at 190°C of 5 to 20 g/10 min, more preferably from 7 to 15 g/10 min.

[00048] Preferably, the polymer composition without any additive, preferably without filler (C) has a melt flow rate MFR₅ according to ISO 1133 at 190°C of 20 to 40 g/10 min, more preferably of 25 to 35 g/10 min.

[00049] Moreover, it is preferred that the melt flow ratio, which is a ratio of two melt flow rates measured for the same polymer under two different loads, falls within a specific range. The preferred specific range is 2.5 to 4.5, more preferably 2.7 to 4.0, for the melt flow ratio MFR₅/MFR₂.

[00050] A further characteristic of the molecular weight distribution (MWD) which is the relation between the number of molecules in a polymer and their individual chain length has to be considered. The width of the distribution is a number as a result of the ratio of the weight average molecular weight divided by the number average molecular weight (M_w/M_n). In the present invention, it is preferred that the polymer composition without any additive, preferably without filler (C), has a M_w/M_n of preferably 8 to 25 and more preferably from 10 to 20.

[00051] Additional additives, e.g. inorganic additives, known as ~~exipients~~ excipients and extrusion aids in the field of coatings and films, are used.

[00052] For a better adhesion between the coating and the substrate, it is preferred that the polymer is oxidized. Consequently, it is preferred that the polymer composition contains anti-oxidants and process stabilizers less than 2,000 ppm, more preferably less than 1,000 ppm and most preferably not more than 700 ppm. The anti-oxidants thereby may be selected from those

known in the art like those containing hindered phenols, secondary aromatic amines, thio-ethers or other sulfur-containing compounds, phosphites and the like including their mixtures.

[00053] It has been found that the polymer composition as described above has a very low water-vapor transmission rate (WVTR). Additionally, the composition has a good adhesion to the substrate, in particular to ~~aluminium~~ aluminum, without any need to have an adhesion layer between the substrate and the coating. Further, the tendency of the coated article to curl is significantly reduced for the polymer composition compared to neat polymer. These advantageous effects could only be achieved as the miscibility between the polymer and the filler is much higher for a multimodal or bimodal polymer having a low molecular weight polymer fraction in comparison with a polymer having the same melt index and density.

[00054] In one preferable embodiment, the multimodal composition comprises as polymer (A), which is the low molecular weight fraction, a polyolefin (1), more preferably a low density polyethylene (LDPE) or linear low density polyethylene (LLDPE). The polyolefin (B), which is the high molecular weight fraction, is a low density polyethylene (LDPE) or a linear low density polyethylene (LLDPE). Preferably, this composition comprises a further polymer (A) which is a wax (2) as defined above. This composition can be produced in an *in situ* process or can be blended mechanically. Preferred properties for the polymer (A), in particular the polyolefin (1), the wax (2) and the polyolefin (B) are those as given above. In case this composition comprises two polymers (A), namely a polyolefin (1) and a wax (2), it is preferred that the amount of wax (2) in the total composition without filler (C) is 1 to 30 wt%, more preferably 1 to 20 wt% and most preferably 1 to 10 wt%. In turn, the composition comprises 70 to 99 wt%, more preferably 80 to 99 wt% and most preferably 90 to 99 wt% of LLDPE resulting from polymer (A) and polyolefin (B). In case the composition comprises LDPE, it is preferred that wax (2) is present in the amount of 1 to 30 wt% and LDPE resulting at least from polymer (B) and optionally from polymer (A) and is present in the amount of 70 to 99 wt% in the total composition without filler (C).

[00055] In another preferable embodiment, a polymer composition is produced in an *in situ* process, whereby the sequential step process by utilizing reactors coupled in series as described above is preferred. Preferably polymer (A) is produced in a loop reactor whereas

polyolefin (B) is produced in a gas phase reactor in the presence of polymer (A). Thereby, it is preferred that the multimodal polymer is at least a bimodal polymer. More preferably, polymer (A) and polyolefin (B) are both polyolefins. The polymer composition of this embodiment comprises 50 to 99 wt% of a linear low density polyethylene (LLDPE) having a multimodal, more preferably a bimodal molecular weight distribution (MWD) and more preferably 1 to 50 wt% of a filler (C), preferably a plate- or sheet-like filler such as mica or talc as described above.

[00056] In the following, when the description refers to LLDPE, it means that a multimodal **polymer**, preferably bimodal LLDPE is used, which comprises a low molecular weight (LMW) fraction, which is polymer (A) (polyolefin (1)), and a high molecular weight (HMW) fraction, which is polymer (B).

[00057] Preferably, the linear low density polyethylene (LLDPE) has a melt index MFR₂ from 1.0 to 20 g/10 min, more preferably from 2 to 15 g/10 min and most preferably from 3 to 10 g/10 min. It is preferred that the linear low density polyethylene (LLDPE) is at below 945 and ranges preferably from 905 to 935 kg/m³, more preferably from 910 to 930 kg/m³, most preferably from 915 to 925 kg/m³. If the melt index of the linear low density polyethylene (LLDPE) is lower than 1 g/10 min, a high throughput is not reached. On the other hand, if the melt index MFR₂ is higher than 20, the melt strength of the polyethylene suffers.

[00058] In addition, it is preferred that the linear low density polyethylene (LLDPE) has a melt flow index MFR₅ from 20 to 40 and preferably a melt flow ratio MFR₅/MFR₂ from 2.5 to 4.5, more preferably from 2.7 to 4.0. Furthermore, it is preferred that the linear low density polyethylene (LLDPE) has a weight average molecular weight (M_w) from 50,000 to 150,000 g/mol, more preferably from 60,000 to 100,000 g/mol and preferably a ratio of the weight average molecular weight to the number average molecular weight M_w/M_n of 8 to 25, more preferably of 10 to 20.

[00059] Moreover, the linear low density polyethylene (LLDPE) contains comonomers selected from the group consisting of C₃ alpha-olefin, C₄ alpha-olefin, C₅ alpha-olefm, C₆ alpha-olefin, C₇ alpha-olefin, C₈ alpha-olefin, C₉ alpha-olefin, C₁₀ alpha-olefin, C₁₁ alpha-olefin, C₁₂ alpha-olefin, C₁₃ alpha-olefin, C₁₄ alpha-olefin, C₁₅ alpha-olefin, C₁₆ alpha-olefin, C₁₇ alpha-olefin, C₁₈ alpha-olefin, C₁₉ alpha-olefin, C₂₀ alpha-olefin. Especially preferred are alpha-olefins

selected from the group consisting of propylene, 1-butene, 1-pentene, 4-methyl-1-pentene, 1-hexene, 1-heptene, 1-octene, 1-nonene, 1-decene, 6-methyl-1-heptene, 4-ethyl-1-hexene, 6-ethyl-1-octene and 7-methyl-1-octene. Still more preferred, alpha-olefins are selected from the group consisting of 1-butene, 4-methyl-1-pentene, 1-hexene and 1-octene.

[00060] As one requirement of the preferred embodiment is that the polymer composition is a linear low density polyethylene (LLDPE) the content of the comonomer units in the polymer is preferably 0.1 to 1.0 mol%, more preferably 0.15 to 0.5 mol%.

[00061] It is preferred that the linear low density polyethylene (LLDPE) without filler (C) comprises 40 to 60 wt%, more preferably 49 to 55 wt% polymer (A) and 60 to 40 wt%, and more preferably 51 to 45 wt% polyolefin (B).

[00062] As stated above, it is preferred that the linear low density polyethylene (LLDPE) comprises a LMW fraction, which forms the polymer (A). More preferably, the polymer (A), is a polyolefin (1), most preferably an ethylene copolymer containing alpha-olefins other than ethylene and listed above. Furthermore, it is preferred that the polymer (A) of the linear low density polyethylene (LLDPE) has a weight average molecular weight (M_w) of 10,000 to 60,000 g/mol, more preferably from 20,000 to 50,000 g/mol. It is further preferred that polymer (A) of the linear high density polyethylene (LLDPE), has a density of at least 905 to 935 kg/m³, more preferably of at least 910 to 930 kg/m³. In addition, it is preferred that polymer (A) of the linear low density polyethylene (LLDPE) has a melt flow rate MFR₂ from 1.0 to 20.0 g/10 min, more preferably from 2.0 to 15.0 g/10 min and most preferred from 3 to 10 g/10 min.

[00063] It is preferred that polyolefin (B) as the linear low density polyethylene (LLDPE) is an ethylene copolymer containing one or more alpha-olefins as listed above. Thereby, it is preferred that the amount of comonomer units in polyolefin (B) is from 2.0 to 15.0 mol%, more preferably from 3.0 to 10.0 mol%. In addition, it is preferred that the polyolefin (B) in the linear low density polyethylene (LLDPE) has a weight average molecular weight from 80,000 to 300,000 g/mol, more preferably from 100,000 to 200,000 g/mol.

[00064] The filler (C) and other additional components in the linear low density polyethylene (LLDPE) are identically used as listed and described above. It is in particular

preferred that additionally to the LLDPE, a wax (2), more preferably a polypropylene wax (2a) or an alkyl-ketene dimer (2b) as defined above is used as an additional polymer (A).

[00065] In case two polymers (A) are used, namely polyolefin (1) and wax (2), the amount of wax (2) is 1 to 30 wt%, more preferably 2 to 20 wt% and most preferably 1 to 10 wt% in the total composition without filler (C). In turn, the composition without filler (C) comprises 70 to 99 wt%, more preferably 80 to 88 wt% and most preferably 90 to 99 wt% LLDPE resulting from polymer (A) and polyolefin (B).

[00066] The further preferable embodiment of the present invention is a polymer composition whereby polymer (A) and polyolefin (B) are preferably mechanically blended. Thereby it is preferred that polymer (A) is a wax (2), more preferably a polypropylene wax (2a) or an alkyl-ketene dimer wax (2b).

[00067] In case of polymer (A), where a polypropylene wax (2a) is used, it is preferred that this wax (2a) has a weight average molecular weight (M_w) of 100 to 50,000, more preferably from 100 to 10,000, and most preferably from 5,000 to 6,000. In addition, it is preferred that the z-average molecular weight of the polypropylene wax (2a) ranges from 100 to 60,000 g/mol, and more preferably from 100 to 10,000 g/mol. It is preferred that the polypropylene wax (2a) has a number average molecular weight (M_n) of 100 to 2,000 g/mol, more preferably 500 to 3,000 g/mol. The melting temperature in DSC-analysis of the polypropylene wax (2a) is preferably of 95 to 130°C, more preferably 105 to 115°C.

[00068] Preferably, the polypropylene wax (2a) is mechanically blended with an ethylene polymer as a polyolefin (B) having an MFR_2 of 6.5 to 8.5 g/10 min, more preferably from 7 to 8 g/10 min and a density of 900 to 940 kg/m³, more preferably from 915 to 925 kg/m³. It is in particular preferred that polyolefin (B) is a low density polyethylene (LDPE), or a linear low density polyethylene (LLDPE) as described above.

[00069] The mechanically blended polymer including a talc as filler (C) and CaO as a water-absorbent component has preferably a density ranging from 1,000 kg/m³ to 1,300 kg/m³, more preferably of 1,150 to 1,200 kg/m³ and a melt flow rate MFR_2 of preferably 8 to 9.5 g/10 min, and more preferably of 8.5 to 9.0 g/10 min.

[00070] The other preferred alternative of a mechanical blend of wax (2) with polyolefin (B) is to use an alkyl-ketene dimer (2b) as wax (2). Preferably, this alkyl-ketene dimer (2b) has a weight average molecular weight (M_w) of 300 to 400 g/mol, more preferably from 320 to 350 g/mol. Preferably, the z-average molecular weight of the alkyl-ketene dimer (2b) is from 300 to 400 g/mol, more preferably from 360 to 390 g/mol. It is preferred that the alkyl-ketene dimer (2b) has a number average molecular weight (M_n) of 200 to 450 g/mol, more preferably from 280 to 300 g/mol. In addition, it is preferred that the alkyl-ketene dimer (2b) has a melting temperature DSC-analysis of 55 to 70°C, more preferably from 60 to 65°C.

[00071] For polyolefin (B), the same ethylene polymer is used as defined under the mechanical blend comprising a polypropylene wax (2a).

[00072] The density of the mechanically blended polymer composition comprising an alkyl-ketene dimer (2b) as defined above, an ethylene polymer (B) as defined above, a filler (C) and a water-absorbent component has preferably a density of 1,050 to 1,300 kg/m³ and more preferably from 1,150 to 1,250 kg/m³. The melt flow rate MFR₂ of this polymer composition is preferably from 12.5 g/10 min to 14.5 g/10 min and more preferably from 13 to 14 g/10 min. It is preferred that for this embodiment for filler (C) talc is employed and for the water absorbent compound CaO.

[00073] Furthermore, the present invention comprises a process for producing the multimodal composition as defined above.

[00074] A multimodal or at least bimodal, e.g. bimodal or trimodal, polymer may be produced by blending two or more monomodal polymers having differently centered maxima in their molecular weight distributions. The blending may be effected mechanically, e.g. analogously to the mechanical blending principles ~~principles~~ known in the art. Alternatively, the multimodal or at least bimodal, e.g. bimodal or trimodal, polymer composition may be produced by polymerization using conditions which create a multimodal or at least bimodal, e.g. bimodal or trimodal, polymer composition, i.e. using a catalyst system for mixtures with two or more different catalytic sides, using two or more stage polymerization process with different process conditions in the different stages (i.e. different temperatures, pressures, polymerization media, hydrogen partial pressures, etc.). With the polymer as produced in such a sequential step

process, i.e. by utilizing reactors coupled in series, and using different conditions in each reactor, the different polymer fractions produced in the different reactors will each have their own molecular weight distribution which may differ considerably from one another. The molecular weight distribution curve of the resulting final polymer can be regarded as superimposing of the molecular weight distribution curves of the polymer fractions which will accordingly show two or more distinct maxima, or at least the distinctively broadened maxima compared with the curves for individual fractions.

[00075] A polymer showing such a molecular weight distribution curve is called multimodal, trimodal or bimodal.

[00076] Multimodal polymers can be produced according to several processes, which are described, e.g. in WO 92/12182 and WO 97/22633.

[00077] A multimodal polymer is preferably produced in a multi-stage process in a **multi-stage** ~~multistage~~ reaction sequence, such as described in WO 92/12182. The contents of this document are included herein by reference.

[00078] It is known to produce multimodal or at least bimodal, e.g. bimodal or trimodal, polymers, preferably multimodal or bimodal olefin-polymers, such as multimodal or bimodal polyethylenes in two or more reactors connected in series whereby the compounds (A) and (B) can be produced in any order.

[00079] According to the present invention, the main polymerization stages are preferably carried out as a combination of a slurry gas/gas-phase polymerization. The slurry polymerization is preferably performed in a so-called loop-reactor.

[00080] Optionally, and of more advantage, the main polymerization stages may be preceded by a pre-polymerization in which case up to 20 wt%, preferably 1-10 wt%, more preferably 1-5 wt% of the total amount of polymer composition is produced. At the pre-polymerization point, all of the catalyst is preferably charged into a loop-reactor and a polymerization is performed as a slurry polymerization. Such a polymerization leads to less fine particles being produced in the following reactors and to a more homogeneous product being obtained in the end. Such a pre-polymerization is for instance described in WO 96/18662.

[00081] Generally, the technique results in a multimodal or at least bimodal, e.g. bimodal or trimodal, polymer composition thereby a Ziegler-Natta or metallocene catalyst in several successive polymerization reactors is used. For example in the production of a bimodal high-density polyethylene composition, a first ethylene polymer is produced in the first reactor under certain conditions with respect to the hydrogen-gas concentration, temperature, pressure and so forth. After the polymerization the reactor-polymer including the catalyst is separated from the reaction mixture and transferred to a second reactor where further polymerization takes place under other conditions.

[00082] The components (A) and (B) can be produced with any suitable catalyst system, preferably a coordination catalyst, such as a Ziegler-Natta catalyst system, preferably a coordination catalyst, such as a Ziegler-Natta catalyst of a transition metal of a group 3-10 of the periodic table (TUPAC), a metallocene, non-metallocene, in a manner known in the art. One example of a preferred Ziegler-Natta catalyst comprises Ti, Mg and Al, such as described in document EP 0 688 794 B1, which is included herewith by reference. It is a high-activity procatalyst comprising a particular inorganic support, a curing compound deposited on the support, wherein the curing compound is the same as or different from the titanium compound, whereby the inorganic support is contacted with an alkyl metal chloride which is soluble in a non-polar hydrocarbon solvent, and has the formula $(R_nMeCl_{3-n})_m$, wherein R is a C_1 to C_{20} alkyl group, Me is a metal of Group III(13) of the periodic table, $n = 1$ or 2 and $m = 1$ or 2 , to give a first reaction product, and the first reaction product is contacted with a compound containing hydrocarbyl and hydrocarbyl oxide linked to magnesium which is soluble in non-polar hydrocarbon solvents, to give a second reaction product, and the second reaction product is contacted with a titanium compound which contains chlorine, having the formula $Cl_xTi(OR^{IV})_{4-x}$, wherein R^{IV} is a C_2 to C_{20} hydrocarbyl group and $x = 3$ or 4 , to give the procatalyst. Preferred supports are inorganic oxides, more preferably silicon dioxide or silica. Most preferably silica having an average particle size of $20\ \mu m$ is used. Even more preferred tri-ethyl aluminium as a ~~co-catalyst~~ co-catalyst is used. Alternatively, a metallocene of group 4 metal can be used.

[00083] Preferably, polymer (A), the low molecular weight (LMW) polymer, is produced with addition or no addition of comonomer in a first reactor, and also the polyolefin (B), the high

molecular weight (HMW) polymer, is produced with addition or no addition, more preferably with addition, of comonomer in the second reactor.

[00084] The resulting end product consists of an intimate mixture of polymers from the two reactors, the different molecular weight distribution occurs of these polymers together forming a molecular weight distribution curve having a broad maximum or two maxima, i.e. the end product is a multimodal or bimodal polymer mixture. Since multimodal and, in particular, bimodal polymers, preferably ethylene polymers and the production thereof belong to the prior art, no detailed description is called for here, but reference is made to the above-mentioned document WO 92/12182. It will be noted that the order of the reaction stages may be reversed.

[00085] Preferably, as stated above, the multimodal polymer composition according to the invention is a bimodal or trimodal polymer composition. It is also preferred that this bimodal or trimodal polymer composition has been produced by polymerization as described above under different polymerization conditions in two or more polymerization reactors connected in series.

[00086] Furthermore, it is preferred that for the multimodal composition according to this invention a process is used as defined above whereby a) polymer (A) and polyolefin (B) are produced together in a multi-stage process comprising a loop reactor and a gas-phase reactor, wherein polymer (A) is generated in at least one loop reactor and the polyolefin (B) is generated in a gas-phase reactor in the presence of the reaction product (A) of the loop reactor, and b) filler (C) and the composition comprising polymer (A) and polyolefin (B) are blended together and compounded.

[00087] In particular, a multi-stage process is used as described above. Especially, it is preferred that a loop reactor is operated at 75 to 100°C, more preferably in the range of 85 to 100°C and most preferably in the range of 90 to 98°C. Thereby, the pressure is preferably 58 to 68 bar, more preferably 60 to 65 bar.

[00088] Preferably, polymer (A) is prepolymerized in a first loop reactor and then continuously removed to a second loop reactor where the polymer (A) is further polymerized. It is preferred that the temperature in the second loop reactor is 90 to 98°C, more preferably about 95°C. Thereby, the pressure is preferably 58 to 68 bar, more preferably about 60 bar.

[00089] In addition, it is preferred that in the second loop reactor, the ethylene concentration is 4 to 10 mol%, more preferably 5 to 8 mol% and most preferably about 6.7 mol%.

[00090] The hydrogen to ethylene mol-ratio highly depends on the catalyst used. It must be adjusted to render the desired melt flow rate MFR of the polymer withdrawn from the loop reactor. For the preferred catalyst as described it is preferred that the ratio of hydrogen to ethylene is 100 to 800 mol/kmol and more preferably 300 to 700 mol/kmol, still more preferably 400 to 650 mol/kmol and most preferred about 550 mol/kmol.

[00091] The polymer slurry is then preferably removed from the loop reactor by using settling tanks and is then preferably introduced into a flash vessel operating preferably at about 3 bar pressure, where the polymer is separated from most of the fluid phase. The polymer is then preferably transferred into a gas-phase reactor operating preferably at 75 to 95°C, more preferably 80 to 90°C and most preferably about 85°C, and at preferably 10 to 50 bar, more preferably 15 to 25 bar and most preferably about 20 bar.

[00092] Additionally, ethylene comonomers were used and hydrogen as well as nitrogen as an inert gas are preferably introduced into the reactor so that the fractional ethylene in the fluidization gas is preferably 1 to 10 mol%, more preferably 1 to 5 mol% and most preferably about 2.5 mol% and the ratio of hydrogen to ethylene is preferably 100 to 400 mol/kmol, more preferably 150 to 300 mol/kmol and most preferably about 210 mol/kmol.

[00093] The comonomer to ethylene ratio has influence on the desired density of the bi-modal polymer. Hence, it is preferred that the ratio of comonomer to ethylene is 20 to 150 mol/kmol, more preferably 50 to 100 mol/kmol and most preferably about 80 mol/kmol. Preferably, after the polymer is withdrawn from the gas-phase reactor and then mixed with further additives as anti-oxidants and/or process stabilizers by blending.

[00094] The polymer mix of polymer (A) and polyolefin (B) is then blended with filler (C) and with any suitable method known in the art. These methods include compounding in a twin-screw extruder, like a counter-rotating twin-screw extruder or a co-rotating twin-screw extruder and compounding in a single-screw extruder.

[00095] In addition, the present invention comprises a new multi-layer material comprising at least a) a substrate as a first layer (I) and b) a multimodal polymer composition as described above as at least one further layer (II). Preferably, the multi-layer material consists of a) a substrate as a first layer (I) and b) a multimodal polymer composition as described above as at least one further layer (II).

[00096] It is further preferred that the multi-layer material is a two-layer or three-layer material consisting of a substrate as a first layer and of a polymer composition for the second and third layer, whereby preferably at least the second layer is a polymer composition as defined above. The layers can of course be in any order. Optionally, this multi-layer material comprises adhesion promoters as tetra-iso-propyl titanate, tetra-stearyl titanate, tetrakis(2-ethylhexyl) titanate, poly(dibutyltitanate).

[00097] Preferably, the substrate is selected from the group consisting of paper, paper-board, ~~aluminium~~ aluminum film and plastic film.

[00098] Preferably, the multi-layer material comprises as a further layer (III) a low density polyethylene (LDPE). Thereby, it is preferred that the low density polyethylene has a density of 900 to 950 kg/m³, more preferably from 915 to 925 kg/m³. In addition, it is preferred that the melt flow rate MFR₂ of the low density polyethylene (LDPE) is of 2.0 to 20.0 g/10 min, more preferably from 3.0 to 10.0 g/10 min.

[00099] Preferably, the coating weight of layer (II) comprising the polymer composition according to the present invention ranges from 5 to 60 g/m² and more preferably from 10 to 45 g/m². Additionally, it is preferred that the layer (III) comprising a low density polyethylene (LDPE) as described above has a coating weight of 0 to 25, more preferably from 3 to 18 g/m².

[000100] The present invention also comprises a film, preferably a cast film, comprising the multimodal polymer composition as described above, more preferably, the film consists of the multimodal polymer composition of the present invention.

[000101] Furthermore, the present invention provides a process for producing a multi-layer material comprising the inventive polymer composition as described above. Thereby, it is preferred that the multimodal polymer composition as described above is applied on a substrate

by a film-coating line comprising an unwind, a wind, a chill roll, and a coating die. Preferably, the speed of the coating line ranges from 50 to 5,000 m/min, more preferably from 100 to 1,500 m/min. The coating may be done as any coating line known in the art. It is preferred to employ a coating line with at least two extruders to make it possible to produce multi-layered coatings with different polymers. It is also possible to have arrangements to treat the polymer melt exiting the die to improve adhesion, e.g. by ozone treatment, corona treatment or flame treatment.

[000102] In addition, the present invention comprises the use of the multimodal polymer composition as defined above for extrusion coating, in particular for extrusion coating producing a multi-layer material as described above.

[000103] Furthermore, the present invention relates to the use of the multimodal polymer composition for films, preferably cast films.

DETAILED DESCRIPTION OF THE INVENTION

[000104] In the following the present invention is demonstrated by means of examples.

Examples

Measurements

[000105] WVTR: Water vapor transmission rate was measured at 90 % relative humidity and 38 °C temperature according to the method ASTM E96.

[000106] Basis weight or coating weight: Basis weight (or coating weight) was determined as follows: Five samples were cut off from the extrusion coated paper parallel in the transverse direction of the line. The size of the samples was 10 cm x 10 cm. The samples were dried in an oven at 105 °C for one hour. The samples were then weighed and the coating weight was calculated as the difference between the basis weight of the coated structure and the basis weight of the substrate. The result was given as a weight of the plastic per square meter.

[000107] Molecular weight averages and molecular weight distribution: Molecular weight averages and molecular weight distribution were determined by ISO 16014, part 2 universal

calibration (narrow MWD polystyrene standards (universal alibration) and a set of 2 x mixed bed + 1 x 10⁷ A Tosohas (JP) columns were used).

[000108] Density: Density was determined according to ISO 1183-1987.

[000109] Melt flow rate or melt index: Melt flow rate (also referred to as melt index) was determined according to ISO 1133, at 190 °C. The load used in the measurement is indicated as a subscript, i.e. MFR₂ denotes the MFR measured under 2.16 kg load.

[000110] Flow rate ratio: Flow rate ratio is a ratio of two melt flow rates measured for the same polymer under two different loads. The loads are indicated as a subscript, i.e., FRR_{5/2} denotes the ratio of MFR₅ to MFR₂.

[000111] Curling: Curling was determined by cutting a circular sample having an area of 100 cm² within two hours after the coating. The sample is then allowed freely to curl at the table for two minutes. The curl is then measured as the difference (in mm) from the table to the curled sheet.

Example 1

[000112] A dry blend of pellets was made of 650 kg of the low density polyethylene CA8200 of 300 kg of a talc filler Finntalc MO5SL, manufactured and sold by Mondo Minerals and 50 kg of Clariant PP6100 PP wax. This dry blend was then compounded and pelletized by using the above-mentioned ZSK70 extruder. The melt temperature during the extrusion was 200°C. The composition was then dried at 60°C for 6 hours to remove the moisture. CA8200 is a low density polyethylene designed for extrusion coating, produced and marketed by Borealis. It is produced by free radical polymerization in a high pressure autoclave process. It has an MFR₂ of 7.5 g/10 min and a density of 920 kg/m³. Clariant PP6100 is a low molecular weight propylene polymer having a number average molecular weight of 2,090 g/mol, weight average molecular weight 5,370 g/mol, z-average molecular weight 10,900 g/mol and melting temperature in DSC analysis 109°C. The composition had a density of 1,195.7 kg/m³ and MFR₂ of 6.1 g/10 min.

Comparative Example 1

[000113] The procedure of Example 1 was repeated, except that the amount of CA8200 was 700 kg and Clariant PP6100 was not used. Moreover, no drying at 60°C was done.

Table 1: Data for compositions containing polyolefin and talc used in cast films.

Example	Composition	MFR ₂ g/10 min	Density 920 kg/m ³
Example 1	LD/PP/talc	NA	NA
Comparative Example 1	LD/ - /talc	NA	NA

Example 2

[000114] The composition of Example 1 was used to make a cast film on Collin laboratory scale cast film line, having a single screw extruder with a screw diameter of 30 mm and length to diameter (L/D) ratio of 30. The line speed was about 10 m/s (from 8.9 to 10.3 m/s), the output about 5 kg/h (from 4.91 to 6.07 kg/h), the die temperature 250°C and melt temperature 245°C. The temperature of the chill roll was about 70°C (68 to 72°C). The data can be found in Table 2. The thickness of the film was 45 µm. The WVTR was 5.0 g/m²/24 h.

Example 3

[000115] The procedure of Example 2 was repeated, except that the thickness of the film was 98 µm. The WVTR was 2.3 g/m²/24 h.

Comparative Example 2

[000116] The procedure of Example 3 was repeated, except that the composition of Comparative Example 1 was used in place of the composition of Example 1. Data can be found in Table 2.

Table 2: Cast film data.

Example	Composition	Thickness μm	WVTR $\text{g/m}^2/24\text{ h}$
Example 2	LD/ PP/ talc	45	5.0
Example 3	LD/ PP/ talc	98	2.3
Comparative Example 2	LD/ - /talc	102	2.7

ABSTRACT

The present invention relates to a polymer composition with good chemical properties and barrier properties being multimodal and comprising a polymer (A) having a weight average molecular weight of lower than 6,000 g/mol and a polyolefin (B) having a higher weight average molecular weight than polymer (A) and a filler (C), whereby a polymer composition without filler (C) has a density of 940 kg/m³ or lower.



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Corneliusstrasse 15
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Nr. der Anmeldung / Application No. / Demande de brevet no

PCT/EP 2005 / 0 0 0 2 2 1

Tag des Eingangs / Date of receipt / Date de réception

12.01.2005 ✓

Zeichen des Anmelders / Vertreter - Applicant / Representative ref. no.
- Référence du demandeur ou du mandataire

K 50 783/7nd ✓

Anmelder / Applicant / Demandeur : **Borealis Technology Oy**

Datum / Date **13.01.05**

Empfangsbescheinigung / Receipt for documents / Récépissé de documents

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The European Patent Office hereby acknowledges the receipt of the following:

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1 ✓

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Description (excluding sequence listing part)
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3 ✓

☐ Prioritätsbeleg(e)
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3 ✓

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3 ✓

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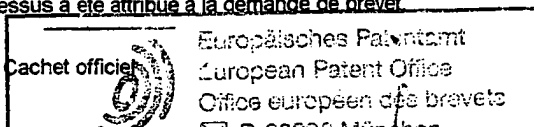
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REQUEST

The undersigned requests that the present international application be processed according to the Patent Cooperation Treaty.

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International Application No.

International Filing Date

Name of receiving Office and "PCT International Application"

Applicant's or agent's file reference
(if desired) (12 characters maximum) K 50 783/7nd

Box No. I TITLE OF INVENTION Extrusion coating polyethylene	
Box No. II APPLICANT <input type="checkbox"/> This person is also inventor	
Name and address: <i>(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)</i> Borealis Technology Oy P.O. Box 330 FIN-06101 Porvoo Finland	Telephone No. Facsimile No. Teleprinter No. Applicant's registration No. with the Office
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Name and address: <i>(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)</i> Kador & Partner Corneliusstr. 15 80469 Munich Germany	Telephone No. +49-89-2015252 Facsimile No. +49-89-2015242 Teleprinter No. Agent's registration No. with the Office
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The filing of this request constitutes under Rule 4.9(a), the designation of all Contracting States bound by the PCT on the international filing date, for the grant of every kind of protection available and, where applicable, for the grant of both regional and national patents.

However,

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Box No. VI PRIORITY CLAIM

The priority of the following earlier application(s) is hereby claimed:

Filing date of earlier application (day/month/year)	Number of earlier application	Where earlier application is:		
		national application: country or Member of WTO	regional application:* regional Office	international application: receiving Office
item (1)				
item (2)				
item (3)				

☐ Further priority claims are indicated in the Supplemental Box.

The receiving Office is requested to prepare and transmit to the International Bureau a certified copy of the earlier application(s) *(only if the earlier application was filed with the Office which for the purposes of this international application is the receiving Office)* identified above as:

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* Where the earlier application is an ARIPO application, indicate at least one country party to the Paris Convention for the Protection of Industrial Property or one Member of the World Trade Organization for which that earlier application was filed (Rule 4.10(b)(ii)):

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Number of
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|---|--|---|
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| <input type="checkbox"/> Box No. VIII (ii) | Declaration as to the applicant's entitlement, as at the international filing date, to apply for and be granted a patent | : |
| <input type="checkbox"/> Box No. VIII (iii) | Declaration as to the applicant's entitlement, as at the international filing date, to claim the priority of the earlier application | : |
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This international application contains:	This international application is accompanied by the following item(s) (mark the applicable check-boxes below and indicate in right column the number of each item):	Number of items
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request (including declaration sheets) : 4	2. <input checked="" type="checkbox"/> original separate power of attorney	: follows
description (excluding sequence listing and/or tables related thereto) : 33	3. <input type="checkbox"/> original general power of attorney	: :
claims : 7	4. <input checked="" type="checkbox"/> copy of general power of attorney; reference number, if any: 42788.	: 1
abstract : 1	5. <input type="checkbox"/> statement explaining lack of signature	: :
drawings : :	6. <input type="checkbox"/> priority document(s) identified in Box No. VI as item(s):	: :
Sub-total number of sheets : 45	7. <input type="checkbox"/> translation of international application into (language):	: :
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Dr. U. Kador

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Annex to the Request

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International Application No.

Applicant's or agent's
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K 50 783/7nd

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Applicant

Borealis Technology Oy

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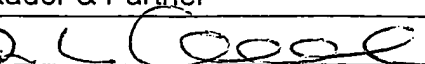
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Date: January 12, 2005

Name: Kador & Partner

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(for several international applications filed under the Patent Cooperation Treaty)

(PCT Rule 90.5)

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(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)

Borealis Technology Oy
P.O. Box 330
FIN-06101 Porvoo
Finland

hereby appoint(s) the following person as:



agent



common representative

Name and address

(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)

Kador & Partner
Corneliusstr. 15
80469 Munich
Germany

to represent the undersigned before



all the competent International Authorities



the International Searching Authority only



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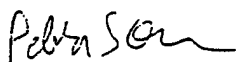
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Pekka Sormunen
Manager Intellectual Property Rights

Date: February 20, 2002

Extrusion coating polyethylene

The present invention relates to a polymer composition suitable for extrusion coating and films, preferably cast films having good chemical properties and barrier properties, in particular, a low water-vapor transmission rate (WVTR) and a low curling. Additionally, the present invention relates to the process for producing the inventive composition and its use. Moreover, the present invention is related to a multi-layer material comprising the polymer composition as well as to a process of said multi-layer material.

One of the largest and most rapidly growing polyolefin-processing method is extrusion coating. The largest single volume of coated materials are different papers and paperboards, which are used for a variety of packaging applications. Other material frequently coated are polymer films, cellophane, aluminium foil, freezer wrap paper and fabrics of various kinds. One target for the improvement of coated articles is to reduce the water-vapor transmission rate (WVTR) as much as possible. A coated material with a low water-vapor transmission rate (WVTR) can for example protect the products wrapped therein much better. The demanded requirement applies, of course, not only to coated materials but also to cast films used for packaging or containers. In both cases, a low water-vapor transmission rate is required. Much effort has been undertaken to improve the water-vapor transmission rate of coated materials as well as for cast films. To date, several new polymer compositions have been developed and much effort has been undertaken to find appropriate fillers to improve the barrier properties significantly. Furthermore, different polymers have been designed as cyclo-olefin copolymers (COC) and liquid crystal polymers (LCP). However, these materials have the drawback of being expensive and having minor processability properties.

WO 00/71615 discloses for example the use of a bimodal high density polyethylene (HDPE) with a melt flow rate, MFR_2 , of 5 g/10 min and a density of 957 kg/m³ for extrusion coating. No information is given how to improve the water-vapor transmission rate (WVTR).

WO 00/34580 describes release liner for pressure-sensitive adhesive labels. The release-liner contains a paper wrap, a filled polymer layer, and, on the opposite of the paper web, an extrudate, e.g. polyethylene, and on the top of the extrudate, a release film. The filled polymer layer can be polyethylene and the filler is an inert particulate, such as silica, mica, clay, talc and titanium oxide. The filler is present in 15 to 40 wt% of the composition.

US 4,978,572 describes a laminated film having three layers. The first layer comprises a thermoplastic resin and 0.3 to 30 wt% white inorganic particles. The second one comprises an ethylene copolymer, 0.5 to 90 wt% of a substance giving anti-block action and anti-oxidant. The third one comprises a metallized thermoplastic. The substance giving anti-block action of the second layer may be silica or talc. The laminated film is reported to have good mechanical strength and good barrier properties.

Even though the prior art offers already a variety of products having good water-vapor transmission rates (WVTR), there is still demand for a significant improvement of these properties. One significant disadvantage in polymer compositions comprising fillers reducing the water-vapor transmission rate (WVTR) is the low dispersion of the fillers incorporated in the polymer matrix. Conventional mechanical incorporation frequently results in poor dispersion as usual fillers form multi-layer aggregation caused by incompatibility with polymer matrix. One consequence of the described phenomenon is that the water-vapor transmission rate (WVTR) varies considerably in the layer leading to unsatisfying average values for the WVTR. Secondly, the low dispersion of the filler causes an

easy upcurling of the polymer composition coated on the materials. Hence, a uniform dispersion of fillers incorporated in a polymer composition should improve the water-vapor transmission rate significantly, and, additionally, the curling properties of a coated material should be enhanced.

Hence, the object of the present invention is to improve the water-vapor transmission rate (WVTR).

The present invention is based on the finding that the object can be addressed by a polymer composition comprising a polymer having a low average molecular weight enabling an enhanced and uniform dispersion of fillers incorporated in the polymer composition.

The present invention therefore provides a multimodal polymer composition comprising

- a) at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60,000 g/mol;
- b) at least one polyolefin (B) having a higher weight average molecular weight (M_w) than polymer (A); and
- c) a filler (C) whereby the polymer composition without filler (C) has a density of 940 kg/m³ or lower.

It is preferred that the polymer composition consists of

- a) at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60,000 g/mol;
- b) at least one polyolefin (B) having a higher weight average molecular weight (M_w) than polymer (A); and
- c) a filler (C) whereby

the polymer composition without filler (C) has a density of 940 kg/m^3 or lower.

Accordingly the polymer composition according to this invention is multimodal with respect to the molecular weight distribution. "Multi-modal" or "multimodal distribution" describes a frequency distribution that has several relative maxima. In particular, the expression "modality of a polymer" refers to the form of its molecular weight distribution (MWD) curve, i.e. the appearance of the graph of the polymer weight fraction as a function of its molecular weight. The molecular weight distribution (MWD) of a polymer produced in a single polymerization stage using a single monomer mixture, a single polymerization catalyst and a single set of process conditions (i.e. temperature, pressure, etc.) shows a single maximum the breadth of which depends on catalyst choice, reactor choice, process conditions, etc., i.e. such a polymer is monomodal.

This inventive composition is characterized by a very low water-vapor transmission rate (WVTR) and also by low curling-values for extrusion-coated layers. These improved properties are reached by a much better dispersion of the filler (C) in the polymer mixture of polymer (A) and polyolefin (B) compared with an unimodal polymer having the same melt index and density for both extrusion-coated layers and cast films.

Hence, the polymer composition according to this invention is a multimodal including bimodal polymer composition consisting of at least two different polymers having two different molecular weight distribution curves and are blended mechanically or in situ during the preparation thereof. Preferably the polymer composition is at least a bimodal mechanical or in-situ blend of a polyolefin (1) (as polymer (A)) and polymer (B). In case such a bimodal blend comprises further a wax (2) as an additional polymer (A), then the final polymer composition may also be trimodal.

The molecular weight distribution (MWD) is the relation between the numbers or molecules in a polymer and their individual chain length. The molecular weight distribution (MWD) is often given as a number, which normally means weight average molecular weight (M_w) divided by number average molecular weight (M_n).

The weight average molecular weight (M_w) is the first moment of a plot of the weight of polymers in each molecular weight range against molecular weight. In turn, the number average molecular weight (M_n) is an average molecular weight of a polymer expressed as the first moment of a plot of the number of molecules in each molecular weight range against the molecular weight. In effect, this is the total molecular weight of all molecules divided by the number of molecules.

The number average molecular weight (M_n) and the weight average molecular weight (M_w) as well as the molecular weight distribution (MWD) are determined according to ISO 16014.

The weight average molecular weight (M_w) is a parameter for the length of the molecules in average. Low M_w -values indicate that the chain length of the molecules is rather short in average. It has been found out that a polymer mixture comprising a polymer (A) with M_w -values of lower than 60,000 g/mol contributes inter alia to better barrier properties and better dispersion of the filler (C). Such better dispersion improves the water-vapor transmission rate (WVTR) as well as the curling resistance positively.

Hence, as a further requirement of the present invention, the multimodal polymer composition must comprise at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60,000 g/mol. It is in particular preferred that at least one polymer (A) having a weight average molecular weight (M_w) of

lower than 60,000 g/mol is at least one polyolefin (1) having a weight average molecular weight (M_w) of 10,000 to 60,000 g/mol, more preferably of 20,000 to 50,000 g/mol and/or at least one wax (2) having a weight average molecular weight (M_w) of less than 10,000 g/mol, more preferably in the range of 500 to 10,000 g/mol.

Moreover, it is preferred that the polyolefin (1) is a polyethylene or polypropylene, more preferably a polyethylene. The polyolefin (1) can be a homopolymer or copolymer. It is preferred that the polyolefin (1) is a homopolymer or copolymer of propylene or ethylene, more preferred the polyolefin (1) is a homopolymer or copolymer of ethylene. Most preferably the polyolefin (1) is a low density polyethylene (LDPE), a linear low density polyethylene (LLDPE), or a linear medium density polyethylene (LMDPE). LDPE, LLDPE and LMDPE are equally suitable alternatives for polyolefin (1), e.g. where a LLDPE or a LMDPE is applicable also a LDPE can be used and vice versa.

In case polymer (A) is a wax (2), it is preferred that it is selected from one or more of

- (2a) a polypropylene wax having a weight average molecular weight (M_w) of less than 10,000 g/mol, more preferably in the range of 500 to 10,000 g/mol, still more preferably in the range of 1000 to 9000 g/mol, yet more preferably in the range of 2000 to 8000 g/mol and most preferably in the range of 4000 to 8000 g/mol or a polyethylene wax having a weight average molecular weight (M_w) of less than 10,000 g/mol, more preferably in the range of 500 to 10,000 g/mol, still more preferably in the range of 1000 to 9000 g/mol, yet more preferably in the range of 2000 to 8000 g/mol and most preferably in the range of 4000 to 8000 g/mol, and
- (2b) an alkyl ketene dimer wax having weight average molecular weight (M_w) of less than 10,000 g/mol, more preferably lower than 5000 g/mol, yet

more preferably lower than 1000 g/mol. In turn the alkyl ketene dimer wax has preferably weight average molecular weight (M_w) of at least 100 g/mol. Most preferred the alkyl ketene dimer wax has weight average molecular weight (M_w) in the range of 250 to 1000 g/mol.

The terms "at least one polymer (A)", "at least one polyolefin (1)" or "at least one wax (2)" shall indicate that more than one polymer (A), polyolefin (1) or wax (2) can be present in the multimodal polymer composition. It is preferred that three, two or one different polymers (A) as defined above are used in a multimodal polymer composition. Still more preferred is that wax (2), preferably a polypropylene wax (2a) or an alkyl ketene dimer wax (2b) as defined above is used as a component (A) only. In case the component (A) comprises a polyolefin (1) as defined above, it is preferred that a wax (2) is present in the multimodal polymer composition as a further polymer (A). In such cases the multimodal composition is preferably trimodal comprising polyolefin (1), wax (2) and polyolefin (B) having different centered maxima in their molecular weight distribution, e.g. having different weight average molecular weights (M_w). The use of the wax (2) has the benefit that the amorphous region of the polymer matrix, which may be a mix of polyolefin (1) and polyolefin (B), is filled up and improves thereby the barrier properties.

It is preferred that not only the final polymer composition has a specific density of 940 kg/m³ or lower but also the polymer (A) shall have a density of lower than 945 kg/m³. It is preferred that polyolefin (1) when used as polymer (A) has a density lower than 945 kg/m³, more preferably is in a range of 905 to 935 kg/m³, still more preferably in the range of 910 to 930 kg/m³ and most preferred in the range of 915 to 925 kg/m³. Preferably, the polyolefin (1) is a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE). In turn, also a low density polyethylene (LDPE) produced in a high pressure process by free radical polymerization is applicable as a polyolefin (1). The linear low

density polyethylene (LLDPE) or the linear medium density polyethylene (LMDPE) is produced by a process as described for the polyolefin (B) below.

The molecular weight distribution (MWD) of the polymer composition is further characterized by the way of its melt flow rate (MFR) according to ISO 1133 at 190°C. The melt flow rate (MFR) mainly depends on the average molecular weight. The reason for this is that long molecules give the material a lower flow tendency than short molecules.

An increase in molecular weight means a decrease in the MFR-value. The melt flow rate (MFR) is measured in g/10 min of the polymer discharged under specific temperature and pressure conditions and is the measure of a viscosity of the polymer which in turn for each type of polymer is mainly influenced by its molecular weight distribution, but also by its degree of branching. The melt flow rate measured under a load of 2.16 kg (ISO 1133) is denoted as MFR_2 . In turn, the melt flow rate measured with 5 kg load (ISO 1133) is denoted as MFR_5 .

In case polymer (A) is a polyolefin (1), it is preferred that MFR_2 is in the range of 1.0 to 20.0 g/10 min and more preferably in the range of 2.0 to 15.0 g/10 min and for example in the range of 3.0 to 10.0 g/10 min. According to one embodiment, the polyolefin (1) is a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE) with MFR_2 as given above. In turn, polyolefin (1) can also be a low density polyethylene (LDPE) having a density in the range as stated in this paragraph. The low density polyethylene (LDPE) is produced in a high-pressure process by free radical polymerization. In turn, the linear low density polyethylene (LLDPE) or the linear medium density polyethylene (LMDPE) is produced as described for polyolefin (B).

In case polymer (A) is an ethylene homopolymer, it is preferred that the ethylene homopolymer contains less than 0.2 mol%, more preferably less than 0.1 mol%

and most preferably less than 0.05 mol% units derived from alpha-olefins other than ethylene. It is in particular preferred that the polymer (A) is an ethylene copolymer, more preferably having a weight average molecular weight from 10,000 to 60,000 g/mol, still more preferably from 20,000 to 50,000 g/mol. Yet more preferably the polymer (A) is an ethylene copolymer having a density of 905 to 935 kg/m³, more preferably of 910 to 930 kg/m³, most preferably of 915 to 925 kg/m³ and having a melt index MFR₂ from 1.0 to 20.0 g/10 min, more preferably from 2.0 to 15.0 g/10 min, most preferably of 3.0 to 10.0 g/10 min. Preferably, the ethylene copolymer comprises, more preferably consists of, comonomer units as defined below for the LLDPE. It is in particular preferred that the ethylene copolymer fulfills all properties as stated above simultaneously.

In case polymer (A) is a wax (2a), namely a polypropylene wax or a polyethylene wax, it is preferred that the wax (2a) has a weight average molecular weight (M_w) in the range of 500 to 10,000 g/mol, more preferably in the range of 1,000 to 9,000 g/mol, still more preferably in the range of 2,000 to 8,000 g/mol and most preferably in the range of 4,000 to 8,000 g/mol. Further preferred ranges for the weight average molecular weight (M_w) of the wax (2a), in particular the polypropylene or polyethylene wax, is in the range of 4,000 to 7,000 g/mol, still more preferably in the range of 5,000 to 6,000 g/mol and most preferably in the range of 5,300 to 5,400 g/mol. Additionally, it is preferred that the wax (2a), in particular the polypropylene wax or polyethylene wax, has a z-average molecular weight of 9,100 to 40,000 g/mol, more preferably from 500 to 20,000 g/mol and most preferably from 10,000 to 12,000 g/mol. It is additionally preferred that the wax (2a), in particular the polypropylene wax or the polyethylene wax, has a number average molecular weight (M_n) of 100 to 20,000 g/mol, more preferably of 500 to 3,000 g/mol.

Moreover, it is preferred that wax (2a), in particular polypropylene wax or polyethylene wax, has a specific molecular weight distribution (MWD) which is the

relation between the number of molecules in the polymer and their individual chain length. The molecular weight distribution is given as a number which means weight average molecular weight divided by number average molecular weight (M_w/M_n). It is preferred that the wax (2a), in particular the polypropylene wax or the polyethylene wax, has an MWD in the range of 1 to 5, more preferably in the range of 1.5 to 4.

In addition, it is preferred that the wax (2a), in particular the polypropylene wax or the polyethylene wax, has a melting temperature in DSC-analysis of below 150°C, more preferably below 140°C, still more preferably in the range of 95 to 130°C, most preferably in a range of 105 to 115°C.

In case a wax (2b), namely an alkyl-ketene dimer, is employed as polymer (A), it is preferred that the weight average molecular weight (M_w) of the wax (2b) is higher than 100 g/mol. In turn, it is preferred that the weight average molecular weight of the wax (2b) is lower than 10,000 g/mol, more preferably lower than 5,000 g/mol, still more preferably lower than 1,000 g/mol. Preferred ranges for the weight average molecular weight (M_w) of the wax (2b) is 100 to 10,000 g/mol, more preferably 250 to 1,000 g/mol. Additionally, it is preferred that the wax (2b) has a number average molecular weight (M_n) of 100 to 20,000 g/mol, more preferably in the range of 100 to 800 g/mol. In addition, it is preferred that wax (2b) has a melting temperature in DSC-analysis below 140°C, more preferably below 100°C. A preferred range for the melting temperature in DSC-analysis is 50 to 90°C, more preferably 50 to 70°C.

As a further requirement, according to the present invention, the polyolefin (B) shall have a higher weight average molecular weight (M_w) than polymer (A). It is preferred that the polyolefin (B) has a weight average molecular weight (M_w) of higher than 80,000 g/mol, more preferably higher than 100,000 g/mol. The upper limit for the weight average molecular weight for polyolefin (B) shall preferably

not be higher than 300,000 g/mol, more preferably not higher than 200,000 g/mol. The preferred range for the weight average molecular weight for polyolefin (B) is 80,000 to 300,000 g/mol, more preferably from 100,000 to 200,000 g/mol. Preferably, polyolefin (B) is a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE), which has been preferably produced in a low medium pressure process in the presence of a polymerization catalyst (i.e. a Ziegler-Natta catalyst or a metallocene catalyst). A linear low density polyethylene (LLDPE) and a linear medium density polyethylene (LMDPE) has a density lower than 945 kg/m^3 , more preferably in the range of 905 to 935 kg/m^3 , still more preferably in the range of 910 to 930 kg/m^3 and most preferred in the range of 915 to 925 kg/m^3 . However, also a low density polyethylene (LDPE) is also applicable for the polyolefin (B). A low density polyethylene (LDPE) has the same density ranges as the LLDPE or the LMDPE as stated in this paragraph and is a product from a high pressure polymerization process characterized by a highly branched chain structure. LDPE, LLDPE and LMDPE are equally suitable alternatives for polyolefin (B), e.g. where a LLDPE or a LMDPE is applicable also a LDPE can be used and vice versa.

According to this invention, more than one polyolefin (B) can be used. Accordingly, the invention also comprises the possibility of any mixture of a linear low density polyethylene (LLDPE), a linear medium density polyethylene (LMDPE) and a low density polyethylene (LDPE).

The MFR₂ of the polyolefin (B) is preferably in the range of 1.0 to 20.0 g/10 min, more preferably in the range of 2.0 to 15.0 g/10 min, in the range of e.g. 3.0 to 10.0 g/10 min. It is in particular preferred that the linear low density polyethylene (LLDPE) and the linear medium density polyethylene (LMDPE) have such melt flow characteristics. In turn, also the low density polyethylene (LDPE) suitable as a polyolefin (B) may have the melt flow characteristics as given in this paragraph.

It is preferred that the polyolefin (B) is a polyethylene. In case the polyolefin (B) is a polyethylene, it may be an ethylene homopolymer or an ethylene copolymer. In case for the polyolefin (B) an ethylene homopolymer is employed, then preferably an ethylene homopolymer is used as defined for polymer (A). In case an ethylene copolymer is employed for polyolefin (B), then preferably an ethylene copolymer is used as defined below. It is in particular preferred that polyolefin (B) is a low density polyethylene (LDPE), a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE).

According to one embodiment, the polymer composition according to this invention is a linear low density polyethylene (LLDPE) comprising polyolefin (1) (polymer (A)) as a low molecular weight fraction of LLDPE and polyolefin (B) as a high molecular weight fraction of LLDPE. This linear low density polyethylene (LLDPE) may be a mechanical blend, preferably an in-situ blend produced in a multistage process. Preferably said composition comprises wax (2) as a further polymer (A).

It is preferred that the polymer composition as defined above comprises 1 to 50 wt% of polymer (A), 40 to 90 wt% of polyolefin (B) and 1 to 50 wt% of filler (C), more preferably of 5 to 40 wt%, and most preferably of 10 to 35 wt%. In case the polymer composition is produced in an in situ polymerization process, e.g. a sequential step process by utilizing reactors coupled in series and described as above, it is preferred that the polymer (A) may range from 40 to 60 wt%, more preferably 49 to 55 wt% in the polymer mix without filler (C). In turn, it is preferred that in such a polymer mix, the polyolefin (B) ranges from 60 to 40 wt%, more preferably from 51 to 45 wt%. Preferably, the total polymer composition comprises 50 to 99 wt% of said polymer mix and of 1 to 50 wt% filler (C), more preferably of 5 to 40 wt%, and most preferably of 10 to 35 wt%.

In case polymer (A) and polyolefin (B) are blended mechanically, it is preferred that polymer (A) ranges from 1 to 30 wt% and, more preferably, from 1 to 20 wt% in the total polymer composition. These ranges apply in particular in case for polymer (A) a wax (2) is used only.

The last requirement according to the present invention is that the multimodal polymer composition additionally comprises a filler (C). Any filler having a positive influence on the water-vapor transmission rate (WVTR) can be used. Preferably, the filler shall be lamellar, such as clay, mica or talc. More preferably, the filler shall be finely divided. The finely divided filler consists of about 95 wt% of particles having particle sizes of less than 10 μm , and about 20-30 wt% of particles having a particle size of less than 1 μm . In the present invention all layer materials may be used as long as they have the ability to disperse in the polymer composition. The filler may either be a clay-based compound or a submicron filler such as talc, calcium carbonate or mica, which usually have been treated, for instance by grinding, to obtain particles of small, i.e. submicron, dimensions, in situ as stated above.

It is preferred that the filler (C) is layered silicate material, still more preferred, filler (C) is a clay-based compound. Clay-based compounds upon compounding of the polymer composition are dispersed in the polymer composition so that individual platelets in the layered structure are separated.

In a further preferred embodiment, the filler (C) is a clay-based layered inorganic, preferably silicate material or material mixture. Such useful clay materials include natural, synthetic and modified phyllosilicates. Natural clays include smectite clays, such as montmorillonite, hectorite, mica, vermiculate, bentonite. Synthetic clays include synthetic mica, synthetic saponite, synthetic hectorite. Modified clays include fluorinated montmorillonite, fluorinated mica.

Of course, the filler (C) may also contain components comprising a mixture of different fillers, such as mixtures of a clay-based filler and talc.

Layered silicates may be made organophylic before being dispersed in the polymer composition by chemical modification, such as by cation-exchange treatment using alkyl ammonium or phosphonium cation complexes. Such cation complexes intercalate between the clay layers.

Preferably, a smectite type clay is used, which comprises montmorillonite, beidellite, nontronite, saponite, as well as hectorite. The most preferred smectite type clay is montmorillonite.

Preferably, also talc is used as a filler (C).

The density affects most physical properties like stiffness impact strength and optical properties of the end products. Hence, and according to the present invention, the density of the polymer composition shall be of 945 kg/m^3 or lower. More preferably, the density shall range from 905 to 935 kg/m^3 , still more preferably from 910 to 930 kg/m^3 and most preferably from 915 to 925 kg/m^3 .

The ranges and values given for the density in the whole invention apply for pure polymer compositions and do not include any additives, in particular no filler (C). The density is determined according to ISO 1183-1987.

Moreover, it is preferred that the polymer composition without any additive, preferably without filler (C) has a melt flow rate MFR_2 according to ISO 1133 at 190°C of 5 to 20 g/10 min , more preferably from 7 to 15 g/10 min .

Preferably, the polymer composition without any additive, preferably without filler (C) has a melt flow rate MFR_5 according to ISO 1133 at 190°C of 20 to 40 g/10 min, more preferably of 25 to 35 g/10 min.

Moreover, it is preferred that the melt flow ratio, which is a ratio of two melt flow rates measured for the same polymer under two different loads, falls within a specific range. The preferred specific range is 2.5 to 4.5, more preferably 2.7 to 4.0, for the melt flow ratio MFR_5/MFR_2 .

A further characteristic of the molecular weight distribution (MWD) which is the relation between the number of molecules in a polymer and their individual chain length has to be considered. The width of the distribution is a number as a result of the ratio of the weight average molecular weight divided by the number average molecular weight (M_w/M_n). In the present invention, it is preferred that the polymer composition without any additive, preferably without filler (C), has a M_w/M_n of preferably 8 to 25 and more preferably from 10 to 20.

Additional additives, e.g. inorganic additives, known as exipients and extrusion aids in the field of coatings and films, are used.

For a better adhesion between the coating and the substrate, it is preferred that the polymer is oxidized. Consequently, it is preferred that the polymer composition contains anti-oxidants and process stabilizers less than 2,000 ppm, more preferably less than 1,000 ppm and most preferably not more than 700 ppm. The anti-oxidants thereby may be selected from those known in the art like those containing hindered phenols, secondary aromatic amines, thio-ethers or other sulfur-containing compounds, phosphites and the like including their mixtures.

It has been found that the polymer composition as described above has a very low water-vapor transmission rate (WVTR). Additionally, the composition has a

good adhesion to the substrate, in particular to aluminium, without any need to have an adhesion layer between the substrate and the coating. Further, the tendency of the coated article to curl is significantly reduced for the polymer composition compared to neat polymer. These advantageous effects could only be achieved as the miscibility between the polymer and the filler is much higher for a multimodal or bimodal polymer having a low molecular weight polymer fraction in comparison with a polymer having the same melt index and density.

In one preferable embodiment, the multimodal composition comprises as polymer (A), which is the low molecular weight fraction, a polyolefin (1), more preferably a low density polyethylene (LDPE) or linear low density polyethylene (LLDPE). The polyolefin (B), which is the high molecular weight fraction, is a low density polyethylene (LDPE) or a linear low density polyethylene (LLDPE). Preferably, this composition comprises a further polymer (A) which is a wax (2) as defined above. This composition can be produced in an in situ process or can be blended mechanically. Preferred properties for the polymer (A), in particular the polyolefin (1), the wax (2) and the polyolefin (B) are those as given above. In case this composition comprises two polymers (A), namely a polyolefin (1) and a wax (2), it is preferred that the amount of wax (2) in the total composition without filler (C) is 1 to 30 wt%, more preferably 1 to 20 wt% and most preferably 1 to 10 wt%. In turn, the composition comprises 70 to 99 wt%, more preferably 80 to 99 wt% and most preferably 90 to 99 wt% of LLDPE resulting from polymer (A) and polyolefin (B). In case the composition comprises LDPE, it is preferred that wax (2) is present in the amount of 1 to 30 wt% and LDPE resulting at least from polymer (B) and optionally from polymer (A) and is present in the amount of 70 to 99 wt% in the total composition without filler (C).

In another preferable embodiment, a polymer composition is produced in an in situ process, whereby the sequential step process by utilizing reactors coupled in series as described above is preferred. Preferably polymer (A) is produced in a

loop reactor whereas polyolefin (B) is produced in a gas phase reactor in the presence of polymer (A). Thereby, it is preferred that the multimodal polymer is at least a bimodal polymer. More preferably, polymer (A) and polyolefin (B) are both polyolefins. The polymer composition of this embodiment comprises 50 to 99 wt% of a linear low density polyethylene (LLDPE) having a multimodal, more preferably a bimodal molecular weight distribution (MWD) and more preferably 1 to 50 wt% of a filler (C), preferably a plate- or sheet-like filler such as mica or talc as described above.

In the following, when the description refers to LLDPE, it means that a multimodal, preferably bimodal LLDPE is used, which comprises a low molecular weight (LMW) fraction, which is polymer (A) (polyolefin (1)), and a high molecular weight (HMW) fraction, which is polymer (B).

Preferably, the linear low density polyethylene (LLDPE) has a melt index MFR_2 from 1.0 to 20 g/10 min, more preferably from 2 to 15 g/10 min and most preferably from 3 to 10 g/10min. It is preferred that the linear low density polyethylene (LLDPE) is at below 945 and ranges preferably from 905 to 935 kg/m³, more preferably from 910 to 930 kg/m³, most preferably from 915 to 925 kg/m³. If the melt index of the linear low density polyethylene (LLDPE) is lower than 1 g/10 min, a high throughput is not reached. On the other hand, if the melt index MFR_2 is higher than 20, the melt strength of the polyethylene suffers.

In addition, it is preferred that the linear low density polyethylene (LLDPE) has a melt flow index MFR_5 from 20 to 40 and preferably a melt flow ratio $\text{MFR}_5/\text{MFR}_2$ from 2.5 to 4.5, more preferably from 2.7 to 4.0. Furthermore, it is preferred that the linear low density polyethylene (LLDPE) has a weight average molecular weight (M_w) from 50,000 to 150,000 g/mol, more preferably from 60,000 to 100,000 g/mol and preferably a ratio of the weight average molecular

weight to the number average molecular weight M_w/M_n of 8 to 25, more preferably of 10 to 20.

Moreover, the linear low density polyethylene (LLDPE) contains comonomers selected from the group consisting of C_3 alpha-olefin, C_4 alpha-olefin, C_5 alpha-olefin, C_6 alpha-olefin, C_7 alpha-olefin, C_8 alpha-olefin, C_9 alpha-olefin, C_{10} alpha-olefin, C_{11} alpha-olefin, C_{12} alpha-olefin, C_{13} alpha-olefin, C_{14} alpha-olefin, C_{15} alpha-olefin, C_{16} alpha-olefin, C_{17} alpha-olefin, C_{18} alpha-olefin, C_{19} alpha-olefin, C_{20} alpha-olefin. Especially preferred are alpha-olefins selected from the group consisting of propylene, 1-butene, 1-pentene, 4-methyl-1-pentene, 1-hexene, 1-heptene, 1-octene, 1-nonene, 1-decene, 6-methyl-1-heptene, 4-ethyl-1-hexene, 6-ethyl-1-octene and 7-methyl-1-octene. Still more preferred, alpha-olefins are selected from the group consisting of 1-butene, 4-methyl-1-pentene, 1-hexene and 1-octene.

As one requirement of the preferred embodiment is that the polymer composition is a linear low density polyethylene (LLDPE) the content of the comonomer units in the polymer is preferably 0.1 to 1.0 mol%, more preferably 0.15 to 0.5 mol%.

It is preferred that the linear low density polyethylene (LLDPE) without filler (C) comprises 40 to 60 wt%, more preferably 49 to 55 wt% polymer (A) and 60 to 40 wt%, and more preferably 51 to 45 wt% polyolefin (B).

As stated above, it is preferred that the linear low density polyethylene (LLDPE) comprises a LMW fraction, which forms the polymer (A). More preferably, the polymer (A), is a polyolefin (1), most preferably an ethylene copolymer containing alpha-olefins other than ethylene and listed above. Furthermore, it is preferred that the polymer (A) of the linear low density polyethylene (LLDPE) has a weight average molecular weight (M_w) of 10,000 to 60,000 g/mol, more preferably from 20,000 to 50,000 g/mol. It is further preferred that polymer (A) of the

linear high density polyethylene (LLDPE), has a density of at least 905 to 935 kg/m³, more preferably of at least 910 to 930 kg/m³. In addition, it is preferred that polymer (A) of the linear low density polyethylene (LLDPE) has a melt flow rate MFR₂ from 1.0 to 20.0 g/10 min, more preferably from 2.0 to 15.0 g/10 min and most preferred from 3 to 10 g/10min.

It is preferred that polyolefin (B) as the linear low density polyethylene (LLDPE) is an ethylene copolymer containing one or more alpha-olefins as listed above. Thereby, it is preferred that the amount of comonomer units in polyolefin (B) is from 2.0 to 15.0mol%, more preferably from 3.0 to 10.0 mol%. In addition, it is preferred that the polyolefin (B) in the linear low density polyethylene (LLDPE) has a weight average molecular weight from 80,000 to 300,000 g/mol, more preferably from 100,000 to 200,000 g/mol.

The filler (C) and other additional components in the linear low density polyethylene (LLDPE), are identically used as listed and described above. It is in particular preferred that additionally to the LLDPE, a wax (2), more preferably a polypropylene wax (2a) or an alkyl-ketene dimer (2b) as defined above is used as an additional polymer (A).

In case two polymers (A) are used, namely polyolefin (1) and wax (2), the amount of wax (2) is 1 to 30 wt%, more preferably 2 to 20 wt% and most preferably 1 to 10 wt% in the total composition without filler (C). In turn, the composition without filler (C) comprises 70 to 99 wt%, more preferably 80 to 88 wt% and most preferably 90 to 99 wt% LLDPE resulting from polymer (A) and polyolefin (B).

The further preferable embodiment of the present invention is a polymer composition whereby polymer (A) and polyolefin (B) are preferably mechanically

blended. Thereby it is preferred that polymer (A) is a wax (2), more preferably a polypropylene wax (2a) or an alkyl-ketene dimer wax (2b).

In case of polymer (A), where a polypropylene wax (2a) is used, it is preferred that this wax (2a) has a weight average molecular weight (M_w) of 100 to 50,000, more preferably from 100 to 10,000, and most preferably from 5,000 to 6,000. In addition, it is preferred that the z-average molecular weight of the polypropylene wax (2a) ranges from 100 to 60,000 g/mol, and more preferably from 100 to 10,000 g/mol. It is preferred that the polypropylene wax (2a) has a number average molecular weight (M_n) of 100 to 2,000 g/mol, more preferably 500 to 3,000 g/mol. The melting temperature in DSC-analysis of the polypropylene wax (2a) is preferably of 95 to 130°C, more preferably 105 to 115°C.

Preferably, the polypropylene wax (2a) is mechanically blended with an ethylene polymer as a polyolefin (B) having an MFR₂ of 6.5 to 8.5 g/10 min, more preferably from 7 to 8 g/10 min and a density of 900 to 940 kg/m³, more preferably from 915 to 925 kg/m³. It is in particular preferred that polyolefin (B) is a low density polyethylene (LDPE), or a linear low density polyethylene (LLDPE) as described above.

The mechanically blended polymer including a talc as filler (C) and CaO as a water-absorbent component has preferably a density ranging from 1,000 kg/m³ to 1,300 kg/m³, more preferably of 1,150 to 1,200 kg/m³ and a melt flow rate MFR₂ of preferably 8 to 9.5 g/10 min, and more preferably of 8.5 to 9.0 g/10 min.

The other preferred alternative of a mechanical blend of wax (2) with polyolefin (B) is to use an alkyl-ketene dimer (2b) as wax (2). Preferably, this alkyl-ketene dimer (2b) has a weight average molecular weight (M_w) of 300 to 400 g/mol, more preferably from 320 to 350 g/mol. Preferably, the z-average molecular weight of the alkyl-ketene dimer (2b) is from 300 to 400 g/mol, more preferably

from 360 to 390 g/mol. It is preferred that the alkyl-ketene dimer (2b) has a number average molecular weight (M_n) of 200 to 450 g/mol, more preferably from 280 to 300 g/mol. In addition, it is preferred that the alkyl-ketene dimer (2b) has a melting temperature DSC-analysis of 55 to 70°C, more preferably from 60 to 65°C.

For polyolefin (B), the same ethylene polymer is used as defined under the mechanical blend comprising a polypropylene wax (2a).

The density of the mechanically blended polymer composition comprising an alkyl-ketene dimer (2b) as defined above, an ethylene polymer (B) as defined above, a filler (C) and a water-absorbent component has preferably a density of 1,050 to 1,300 kg/m³ and more preferably from 1,150 to 1,250 kg/m³. The melt flow rate MFR₂ of this polymer composition is preferably from 12.5 g/10 min to 14.5 g/10 min and more preferably from 13 to 14 g/10 min. It is preferred that for this embodiment for filler (C) talc is employed and for the water absorbent compound CaO.

Furthermore, the present invention comprises a process for producing the multimodal composition as defined above.

A multimodal or at least bimodal, e.g. bimodal or trimodal, polymer may be produced by blending two or more monomodal polymers having differently centered maxima in their molecular weight distributions. The blending may be effected mechanically, e.g. analogously to the mechanical blending principles known in the art. Alternatively, the multimodal or at least bimodal, e.g. bimodal or trimodal, polymer composition may be produced by polymerization using conditions which create a multimodal or at least bimodal, e.g. bimodal or trimodal, polymer composition, i.e. using a catalyst system for mixtures with two or more different catalytic sides, using two or more stage polymerization process with different

process conditions in the different stages (i.e. different temperatures, pressures, polymerization media, hydrogen partial pressures, etc.). With the polymer as produced in such a sequential step process, i.e. by utilizing reactors coupled in series, and using different conditions in each reactor, the different polymer fractions produced in the different reactors will each have their own molecular weight distribution which may differ considerably from one another. The molecular weight distribution curve of the resulting final polymer can be regarded as superimposing of the molecular weight distribution curves of the polymer fractions which will accordingly show two or more distinct maxima, or at least the distinctively broadened maxima compared with the curves for individual fractions.

A polymer showing such a molecular weight distribution curve is called multimodal, trimodal or bimodal.

Multimodal polymers can be produced according to several processes, which are described, e.g. in WO 92/12182 and WO 97/22633.

A multimodal polymer is preferably produced in a multi-stage process in a multi-stage reaction sequence, such as described in WO 92/12182. The contents of this document are included herein by reference.

It is known to produce multimodal or at least bimodal, e.g. bimodal or trimodal, polymers, preferably multimodal or bimodal olefin-polymers, such as multimodal or bimodal polyethylenes in two or more reactors connected in series whereby the compounds (A) and (B) can be produced in any order.

According to the present invention, the main polymerization stages are preferably carried out as a combination of a slurry gas/gas-phase polymerization. The slurry polymerization is preferably performed in a so-called loop-reactor.

Optionally, and of more advantage, the main polymerization stages may be preceded by a pre-polymerization in which case up to 20 wt%, preferably 1-10 wt%, more preferably 1-5 wt% of the total amount of polymer composition is produced. At the pre-polymerization point, all of the catalyst is preferably charged into a loop-reactor and a polymerization is performed as a slurry polymerization. Such a polymerization leads to less fine particles being produced in the following reactors and to a more homogeneous product being obtained in the end. Such a pre-polymerization is for instance described in WO 96/18662.

Generally, the technique results in a multimodal or at least bimodal, e.g. bimodal or trimodal, polymer composition thereby a Ziegler-Natta or metallocene catalyst in several successive polymerization reactors is used. For example in the production of a bimodal high-density polyethylene composition, a first ethylene polymer is produced in the first reactor under certain conditions with respect to the hydrogen-gas concentration, temperature, pressure and so forth. After the polymerization the reactor-polymer including the catalyst is separated from the reaction mixture and transferred to a second reactor where further polymerization takes place under other conditions.

The components (A) and (B) can be produced with any suitable catalyst system, preferably a coordination catalyst, such as a Ziegler-Natta catalyst system, preferably a coordination catalyst, such as a Ziegler-Natta catalyst of a transition metal of a group 3-10 of the periodic table (IUPAC), a metallocene, non-metallocene, in a manner known in the art. One example of a preferred Ziegler-Natta catalyst comprises Ti, Mg and Al, such as described in document EP 0 688 794 B1, which is included herewith by reference. It is a high-activity pro-catalyst comprising a particular inorganic support, a curing compound deposited on the support, wherein the curing compound is the same as or different from the titanium compound, whereby the inorganic support is contacted with an alkyl metal

chloride which is soluble in a non-polar hydrocarbon solvent, and has the formula $(R_nMeCl_{3-n})_m$, wherein R is a C_1 to C_{20} alkyl group, Me is a metal of Group III(13) of the periodic table, $n = 1$ or 2 and $m = 1$ or 2 , to give a first reaction product, and the first reaction product is contacted with a compound containing hydrocarbyl and hydrocarbyl oxide linked to magnesium which is soluble in non-polar hydrocarbon solvents, to give a second reaction product, and the second reaction product is contacted with a titanium compound which contains chlorine, having the formula $Cl_xTi(OR^{IV})_{4-x}$, wherein R^{IV} is a C_2 to C_{20} hydrocarbyl group and $x = 3$ or 4 , to give the procatalyst. Preferred supports are inorganic oxides, more preferably silicon dioxide or silica. Most preferably silica having an average particle size of $20\ \mu m$ is used. Even more preferred tri-ethyl aluminium as a cocatalyst is used. Alternatively, a metallocene of group 4 metal can be used.

Preferably, polymer (A), the low molecular weight (LMW) polymer, is produced with addition or no addition of comonomer in a first reactor, and also the polyolefin (B), the high molecular weight (HMW) polymer, is produced with addition or no addition, more preferably with addition, of comonomer in the second reactor.

The resulting end product consists of an intimate mixture of polymers from the two reactors, the different molecular weight distribution occurs of these polymers together forming a molecular weight distribution curve having a broad maximum or two maxima, i.e. the end product is a multimodal or bimodal polymer mixture. Since multimodal and, in particular, bimodal polymers, preferably ethylene polymers and the production thereof belong to the prior art, no detailed description is called for here, but reference is made to the above-mentioned document WO 92/12182. It will be noted that the order of the reaction stages may be reversed.

Preferably, as stated above, the multimodal polymer composition according to the invention is a bimodal or trimodal polymer composition. It is also preferred that this bimodal or trimodal polymer composition has been produced by polymerization as described above under different polymerization conditions in two or more polymerization reactors connected in series.

Furthermore, it is preferred that for the multimodal composition according to this invention a process is used as defined above whereby

- a) polymer (A) and polyolefin (B) are produced together in a multi-stage process comprising a loop reactor and a gas-phase reactor, wherein polymer (A) is generated in at least one loop reactor and the polyolefin (B) is generated in a gas-phase reactor in the presence of the reaction product (A) of the loop reactor, and
- b) filler (C) and the composition comprising polymer (A) and polyolefin (B) are blended together and compounded.

In particular, a multi-stage process is used as described above. Especially, it is preferred that a loop reactor is operated at 75 to 100°C, more preferably in the range of 85 to 100°C and most preferably in the range of 90 to 98°C. Thereby, the pressure is preferably 58 to 68 bar, more preferably 60 to 65 bar.

Preferably, polymer (A) is prepolymerized in a first loop reactor and then continuously removed to a second loop reactor where the polymer (A) is further polymerized. It is preferred that the temperature in the second loop reactor is 90 to 98°C, more preferably about 95°C. Thereby, the pressure is preferably 58 to 68 bar, more preferably about 60 bar.

In addition, it is preferred that in the second loop reactor, the ethylene concentration is 4 to 10 mol%, more preferably 5 to 8 mol% and most preferably about 6.7 mol%.

The hydrogen to ethylene mol-ratio highly depends on the catalyst used. It must be adjusted to render the desired melt flow rate MFR of the polymer withdrawn from the loop reactor. For the preferred catalyst as described it is preferred that the ratio of hydrogen to ethylene is 100 to 800 mol/kmol and more preferably 300 to 700 mol/kmol, still more preferably 400 to 650 mol/kmol and most preferred about 550 mol/kmol.

The polymer slurry is then preferably removed from the loop reactor by using settling tanks and is then preferably introduced into a flash vessel operating preferably at about 3 bar pressure, where the polymer is separated from most of the fluid phase. The polymer is then preferably transferred into a gas-phase reactor operating preferably at 75 to 95°C, more preferably 80 to 90°C and most preferably about 85°C, and at preferably 10 to 50 bar, more preferably 15 to 25 bar and most preferably about 20 bar.

Additionally, ethylene comonomers were used and hydrogen as well as nitrogen as an inert gas are preferably introduced into the reactor so that the fractional ethylene in the fluidization gas is preferably 1 to 10 mol%, more preferably 1 to 5 mol% and most preferably about 2.5 mol% and the ratio of hydrogen to ethylene is preferably 100 to 400 mol/kmol, more preferably 150 to 300 mol/kmol and most preferably about 210 mol/kmol.

The comonomer to ethylene ratio has influence on the desired density of the bimodal polymer. Hence, it is preferred that the ratio of comonomer to ethylene is 20 to 150 mol/kmol, more preferably 50 to 100 mol/kmol and most preferably about 80 mol/kmol. Preferably, after the polymer is withdrawn from the gas-

phase reactor and then mixed with further additives as anti-oxidants and/or process stabilizers by blending.

The polymer mix of polymer (A) and polyolefin (B) is then blended with filler (C) and with any suitable method known in the art. These methods include compounding in a twin-screw extruder, like a counter-rotating twin-screw extruder or a co-rotating twin-screw extruder and compounding in a single-screw extruder.

In addition, the present invention comprises a new multi-layer material comprising at least

- a) a substrate as a first layer (I) and
- b) a multimodal polymer composition as described above as at least one further layer (II).

Preferably, the multi-layer material consists of

- a) a substrate as a first layer (I) and
- b) a multimodal polymer composition as described above as at least one further layer (II).

It is further preferred that the multi-layer material is a two-layer or three-layer material consisting of a substrate as a first layer and of a polymer composition for the second and third layer, whereby preferably at least the second layer is a polymer composition as defined above. The layers can of course be in any order. Optionally, this multi-layer material comprises adhesion promoters as tetra-isopropyl titanate, tetra-stearyl titanate, tetrakis(2-ethylhexyl) titanate, poly(dibutyltitanate).

Preferably, the substrate is selected from the group consisting of paper, paper-board, aluminium film and plastic film.

Preferably, the multi-layer material comprises as a further layer (III) a low density polyethylene (LDPE). Thereby, it is preferred that the low density polyethylene has a density of 900 to 950 kg/m³, more preferably from 915 to 925 kg/m³. In addition, it is preferred that the melt flow rate MFR₂ of the low density polyethylene (LDPE) is of 2.0 to 20.0 g/10 min, more preferably from 3.0 to 10.0 g/10 min.

Preferably, the coating weight of layer (II) comprising the polymer composition according to the present invention ranges from 5 to 60 g/m² and more preferably from 10 to 45 g/m². Additionally, it is preferred that the layer (III) comprising a low density polyethylene (LDPE) as described above has a coating weight of 0 to 25, more preferably from 3 to 18 g/m².

The present invention also comprises a film, preferably a cast film, comprising the multimodal polymer composition as described above, more preferably, the film consists of the multimodal polymer composition of the present invention.

Furthermore, the present invention provides a process for producing a multi-layer material comprising the inventive polymer composition as described above. Thereby, it is preferred that the multimodal polymer composition as described above is applied on a substrate by a film-coating line comprising an unwind, a wind, a chill roll and a coating die. Preferably, the speed of the coating line ranges from 50 to 5000 m/min, more preferably from 100 to 1500 m/min. The coating may be done as any coating line known in the art. It is preferred to employ a coating line with at least two extruders to make it possible to produce multilayered coatings with different polymers. It is also possible to have arrange-

ments to treat the polymer melt exiting the die to improve adhesion, e.g. by ozone treatment, corona treatment or flame treatment.

In addition, the present invention comprises the use of the multimodal polymer composition as defined above for extrusion coating, in particular for extrusion coating producing a multi-layer material as described above.

Furthermore, the present invention relates to the use of the multimodal polymer composition for films, preferably cast films.

In the following the present invention is demonstrated by means of examples.

Examples

Measurements

WVTR:

Water vapor transmission rate was measured at 90 % relative humidity and 38 °C temperature according to the method ASTM E96.

Basis weight or coating weight:

Basis weight (or coating weight) was determined as follows: Five samples were cut off from the extrusion coated paper parallel in the transverse direction of the line. The size of the samples was 10 cm × 10 cm. The samples were dried in an oven at 105 °C for one hour. The samples were then weighed and the coating weight was calculated as the difference between the basis weight of the coated structure and the basis weight of the substrate. The result was given as a weight of the plastic per square meter.

Molecular weight averages and molecular weight distribution:

Molecular weight averages and molecular weight distribution were determined by ISO 16014, part 2 universal calibration (narrow MWD polystyrene standards (universal alibration) and a set of 2 x mixed bed + 1 x 10⁷ Å Tosohas (JP) columns were used).

Density:

Density was determined according to ISO 1183-1987.

Melt flow rate or melt index:

Melt flow rate (also referred to as melt index) was determined according to ISO 1133, at 190 °C. The load used in the measurement is indicated as a subscript, i.e. MFR₂ denotes the MFR measured under 2.16 kg load.

Flow rate ratio:

Flow rate ratio is a ratio of two melt flow rates measured for the same polymer under two different loads. The loads are indicated as a subscript, i.e., FRR_{5/2} denotes the ratio of MFR₅ to MFR₂.

Curling:

Curling was determined by cutting a circular sample having an area of 100 cm² within two hours after the coating. The sample is then allowed freely to curl at the table for two minutes. The curl is then measured as the difference (in mm) from the table to the curled sheet.

Example 1

A dry blend of pellets was made of 650 kg of the low density polyethylene CA8200 of 300 kg of a talc filler Finntalc MO5SL, manufactured and sold by Mondo Minerals and 50 kg of Clariant PP6100 PP wax. This dry blend was then compounded and pelletized by using the above-mentioned ZSK70 extruder. The melt temperature during the extrusion was 200°C. The composition was then dried at 60°C for 6 hours to remove the moisture. CA8200 is a low density polyethylene designed for extrusion coating, produced and marketed by Borealis. It is produced by free radical polymerization in a high pressure autoclave process. It has an MFR₂ of 7.5 g/10 min and a density of 920 kg/m³. Clariant PP6100 is a low molecular weight propylene polymer having a number average molecular weight of 2,090 g/mol, weight average molecular weight 5,370 g/mol, z-average molecular weight 10,900 g/mol and melting temperature in DSC analysis 109°C. The composition had a density of 1,195.7 kg/m³ and MFR₂ of 6.1 g/10 min.

Comparative Example 1

The procedure of Example 1 was repeated, except that the amount of CA8200 was 700 kg and Clariant PP6100 was not used. Moreover, no drying at 60°C was done.

Table 1: Data for compositions containing polyolefin and talc used in cast films.

Example	Composition	MFR ₂ g/10 min	Density 920 kg/m ³
Example 1	LD/PP/talc	NA	NA
Comparative Example 1	LD/ - /talc	NA	NA

Example 2

The composition of Example 1 was used to make a cast film on Collin laboratory scale cast film line, having a single screw extruder with a screw diameter of 30 mm and length to diameter (L/D) ratio of 30. The line speed was about 10 m/s (from 8.9 to 10.3 m/s), the output about 5 kg/h (from 4.91 to 6.07 kg/h), the die temperature 250°C and melt temperature 245°C. The temperature of the chill roll was about 70°C (68 to 72°C). The data can be found in Table 2.

The thickness of the film was 45 μm . The WVTR was 5.0 $\text{g}/\text{m}^2/24 \text{ h}$.

Example 3

The procedure of Example 2 was repeated, except that the thickness of the film was 98 μm . The WVTR was 2.3 $\text{g}/\text{m}^2/24 \text{ h}$.

Comparative Example 2

The procedure of Example 3 was repeated, except that the composition of Comparative Example 1 was used in place of the composition of Example 1. Data can be found in Table 2.

Table 2: Cast film data.

Example	Composition	Thickness μm	WVTR $\text{g/m}^2/24 \text{ h}$
Example 2	LD/ PP/ talc	45	5.0
Example 3	LD/ PP/ talc	98	2.3
Comparative Example 2	LD/ - / talc	102	2.7

Claims

1. A multimodal polymer composition comprising
 - a. at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60000 g/mol;
 - b. at least one polyolefin (B) having a higher weight average molecular weight (M_w) than polymer (A); and
 - c. a filler (C), whereby
the polymer composition without filler (C) has a density of 940 kg/m³ or lower.
2. A polymer composition according to claim 1 characterized in that at least one polymer (A) is
 - (1) a polyolefin having a weight average molecular weight (M_w) of 10000 to 60000 g/mol, or
 - (2) a wax having weight average molecular weight (M_w) of less than 10000 g/mol.
3. A polymer composition according to claim 2 characterized in that the polyolefin (1) is a low density polyethylene (LDPE), a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE).
4. A polymer composition according to claim 2 or 3 characterized in that the wax (2) is selected from one or more of
 - (2a) a polypropylene wax having weight average molecular weight (M_w) of less than 10000 g/mol or a polypropylene wax having weight average

molecular weight (M_w) of less than 10000 g/mol, or

(2b) an alkyl ketene dimer wax having weight average molecular weight (M_w) of less than 10000 g/mol.

5. A polymer composition according to any one of the preceding claims 2 to 4 characterized in that the composition comprises a polyolefin (1) as polymer (A) and a wax (2) as a further polymer (A).
6. A polymer composition according to any one of the preceding claims 1 to 5 characterized in that the polymer (A) has a density of lower than 945 kg/m³.
7. A polymer composition according to any one of the preceding claims 1 to 6 characterized in that the multimodal polymer composition is at least a bimodal polymer composition.
8. A polymer composition according to any one of the preceding claims 1 to 7 characterized in that the polyolefin (B) has a weight average molecular weight (M_w) of higher than 80000 g/mol.
9. A polymer composition according to any one of the preceding claims 1 to 8 characterized in that the polyolefin (B) is a polyethylene.
10. A polymer composition according to claim 9 characterized in that the polyolefin (B) is a low density polyethylene (LDPE), a linear low density polyethylene (LLDPE) or linear medium density polyethylene (LMDPE).

11. A polymer composition according to any one of the preceding claims 1 to 10 characterized in that the total polymer composition comprises 1 to 50 wt% of polymer (A), 40 to 90 wt% of polyolefin (B) and 1 to 50 wt% of filler (C).
12. A polymer composition according to any one of the preceding claims 1 to 11 characterized in that the polymer composition without filler (C) has melt flow rate MFR_2 , according to ISO 1133, at 190 °C, of 5 to 20 g/10min.
13. A polymer composition according to any one of the preceding claims 1 to 12 characterized in that the polymer composition without filler (C) has melt flow rate MFR_5 , according to ISO 1133, at 190 °C, of 20 to 40 g/10min.
14. A polymer composition according to any one of the preceding claims 1 to 13 characterized in that the polymer composition without filler (C) has melt flow ratio MFR_5/MFR_2 of 2.5 to 4.5.
15. A polymer composition according to any one of the preceding claims 1 to 14 characterized in that the polymer composition without filler (C) has a ratio of the weight average molecular weight (M_w) to the number average molecular weight (M_n) from 8 to 25.
16. A polymer composition according to any one of the preceding claims 1 to 15 characterized in that 95 wt% of the filler (C) has a particle size of less than 10 μm .

17. A polymer composition according to any one of the preceding claims 1 to 16 characterized in that the filler (C) is talc.
18. A polymer composition according to any one of the preceding claims 1 to 17 characterized in that the polymer composition comprises additionally antioxidants(s) and/or process stabilizers of less than 2000 ppm in the total composition.
19. A polymer composition according to any one of the preceding claims 1 to 18 characterized in that the polymer composition is a linear low density polyethylene (LLDPE) or a liner medium density polyethylene (LMDPE), whereby polymer (A) and polyolefin (B) are produced in a multi step polymerization process.
20. A polymer composition according to claim 19 characterized in that the amount of comonomer units in a linear low density polyethylene (LLDPE) or a in liner medium density polyethylene (LMDPE) is 0.1 to 1.0 mol %.
21. A polymer composition according to claim 19 or 20 characterized in that the polymer (A) and the polyolefin (B) is a linear low density polyethylene (LLDPE) or a liner medium density polyethylene (LMDPE), whereby the comonomer units are selected from the group consisting of C₃ α -olefin, C₄ α -olefin, C₅ α -olefin, C₆ α -olefin, C₇ α -olefin, C₈ α -olefin, C₉ α -olefin, C₁₀ α -olefin, C₁₁ α -olefin, C₁₂ α -olefin, C₁₃ α -olefin, C₁₄ α -olefin, C₁₅ α -olefin, C₁₆ α -olefin, C₁₇ α -olefin, C₁₈ α -olefin, C₁₉ α -olefin and C₂₀ α -olefin.

22. A polymer composition according to any one of the preceding claims 1 to 18 characterized in that the polymer (A) is a wax (2) according to claim (4) and the polyolefin (B) is a linear low density polyethylene (LLDPE) or low density polyethylene (LDPE).
23. A polymer composition according to claim 22 characterized in that the polymer composition comprises additionally a polyolefin (1) being a linear low density polyethylene (LLDPE) as a further polymer (A).
24. A polymer composition according to claim 20 or 21 characterized in that the polymer composition is a linear low density polyethylene (LLDPE) whereby polyolefin (1) (polymer (A)) being a linear low density polyethylene (LLDPE) is the lower molecular weight fraction of LLDPE and polyolefin (B) being a linear low density polyethylene (LLDPE) is the higher molecular weight fraction of the LLDPE.
25. A polymer composition according to claim 24 characterized in that the polymer (A) and polyolefin (B) are a mechanical blend, preferably an in-situ blend produced in a multistage polymerization process.
26. A multi-layer material comprising
 - a. a substrate as a first layer (I)
 - b. a multimodal polymer composition according to any one of the preceding claims as at least a further layer (II).

27. A multi-layer material according to claim 26 characterized in that the substrate is selected from the group consisting of paper, paperboard, aluminum film and plastic film.
28. A multi-layer material according to claim 26 or 27 characterized in that the multi-layer material comprises as a further layer (III) comprising a low density polyethylene (LDPE).
29. A multi-layer material according to any one of the preceding claims 26 to 28 characterized in that the low density polyethylene (LDPE) layer (III) has a melt flow rate MFR_2 , according to ISO 1133, at 190°C, of at least 5 g/10 min.
30. A film comprising a multimodal polymer composition according to any one of the preceding claims 1 to 25.
31. A process for producing a composition according to any one of the preceding claims 1 to 25 characterized in that
 - a. polymer (A) and polyolefin (B) are produced together in a multi-stage process comprising a loop reactor and a gas phase reactor, wherein polymer (A) is generated in at least one loop reactor and the polyolefin (B) is generated in a gas phase reactor; and
 - b. filler (C) and the composition comprising polymer (A) and polyolefin (B) are blended together and compounded.
32. A process for producing a composition according to claim 31 characterized in that the catalyst used for the process producing the composition comprising polymer (A) and polyolefin (B) is a high activity procatalyst comprising a particulate inorganic support, a chlorine compound deposited on the support, wherein the chlorine compound is the same as or different from the titanium compound, whereby the inorganic support is contacted with an alkyl metal chloride which is

soluble in non-polar hydrocarbon solvents, and has the formula $R_n\text{MECL}_3$.
 $n)_m$ wherein R is a $\text{C}_1\text{-C}_{20}$ alkyl group, Me is a metal of group III(13) of
 the periodic table, $n=1$ or 2 and $m=1$ or 2, to give a first reaction product,
 and

the first reaction product is contacted with a compound containing hydro-
 carbyl and hydrocarbyl oxide linked to magnesium which is soluble in
 non-polar hydrocarbon solvents, to give a second reaction product, and
 the first reaction product is contacted with a compound containing hydro-
 carbyl and hydrocarbyl oxide linked to magnesium which is soluble in
 non-polar hydrocarbon solvents, to give a second reaction product, and
 the second reaction product is contacted with a titanium compound which
 contains chlorine, having the formula $\text{Cl}_x\text{Ti}(\text{OR}^{\text{IV}})_{4-x}$ wherein R^{IV} is a $\text{C}_2\text{-}$
 C_{20} hydrocarbyl group and x is 3 or 4, to give the procatalyst.

33. A process for producing a multi-layer material according to any one of the
 claims 26 to 29 characterized in that the multimodal polymer composition
 according to any one of the claims 1 to 25 is applied on the substrate by a
 film coating line comprising an unwind, a wind, a chill roll and a coating
 die.
34. Use of the multimodal polymer composition according to any one of the
 claims 1 to 25 for extrusion coating.
35. Use according to claim 34 characterized in that the polymer extrusion
 composition according to any one of the claims 1 to 25 is used for extru-
 sion coating producing a multi-layer material according to any one of the
 claim 26 to 29.
36. Use of the multimodal polymer composition according to any one of the
 claims 1 to 25 for a film, preferably for a cast film.

Abstract

The present invention relates to a polymer composition with good chemical properties and barrier properties being multimodal and comprising a polymer (A) having a weight average molecular weight of lower than 6000 g/mol and a polyolefin (B) having a higher weight average molecular weight than polymer (A) and a filler (C), whereby a polymer composition without filler (C) has a density of 940 kg/m³ or lower.

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(54) Title: EXTRUSION COATING POLYETHYLENE

(57) Abstract: The present invention relates to a polymer composition with good chemical properties and barrier properties being multimodal and comprising a polymer (A) having a weight average molecular weight of lower than 6000 g/mol and a polyolefine (B) having a higher weight average molecular weight than polymer (A) and a filler (C), whereby a polymer composition without filter (C) has a density of 940 kg/m³ or lower.



WO 2006/074694 A1

Extrusion coating polyethylene

The present invention relates to a polymer composition suitable for extrusion coating and films, preferably cast films having good chemical properties and barrier properties, in particular, a low water-vapor transmission rate (WVTR) and a low curling. Additionally, the present invention relates to the process for producing the inventive composition and its use. Moreover, the present invention is related to a multi-layer material comprising the polymer composition as well as to a process of said multi-layer material.

One of the largest and most rapidly growing polyolefin-processing method is extrusion coating. The largest single volume of coated materials are different papers and paperboards, which are used for a variety of packaging applications. Other material frequently coated are polymer films, cellophane, aluminium foil, freezer wrap paper and fabrics of various kinds. One target for the improvement of coated articles is to reduce the water-vapor transmission rate (WVTR) as much as possible. A coated material with a low water-vapor transmission rate (WVTR) can for example protect the products wrapped therein much better. The demanded requirement applies, of course, not only to coated materials but also to cast films used for packaging or containers. In both cases, a low water-vapor transmission rate is required. Much effort has been undertaken to improve the water-vapor transmission rate of coated materials as well as for cast films. To date, several new polymer compositions have been developed and much effort has been undertaken to find appropriate fillers to improve the barrier properties significantly. Furthermore, different polymers have been designed as cyclo-olefin copolymers (COC) and liquid crystal polymers (LCP). However, these materials have the drawback of being expansive and having minor processability properties.

WO 00/71615 discloses for example the use of a bimodal high density polyethylene (HDPE) with a melt flow rate, MFR₂, of 5 g/10 min and a density of 957 kg/m³ for extrusion coating. No information is given how to improve the water-vapor transmission rate (WVTR).

WO 00/34580 describes release liner for pressure-sensitive adhesive labels. The release-liner contains a paper wrap, a filled polymer layer, and, on the opposite of the paper web, an extrudate, e.g. polyethylene, and on the top of the extrudate, a release film. The filled polymer layer can be polyethylene and the filler is an inert particulate, such as silica, mica, clay, talc and titanium oxide. The filler is present in 15 to 40 wt% of the composition.

US 4,978,572 describes a laminated film having three layers. The first layer comprises a thermoplastic resin and 0.3 to 30 wt% white inorganic particles. The second one comprises an ethylene copolymer, 0.5 to 90 wt% of a substance giving anti-block action and anti-oxidant. The third one comprises a metallized thermoplastic. The substance giving anti-block action of the second layer may be silica or talc. The laminated film is reported to have good mechanical strength and good barrier properties.

Even though the prior art offers already a variety of products having good water-vapor transmission rates (WVTR), there is still demand for a significant improvement of these properties. One significant disadvantage in polymer compositions comprising fillers reducing the water-vapor transmission rate (WVTR) is the low dispersion of the fillers incorporated in the polymer matrix. Conventional mechanical incorporation frequently results in poor dispersion as usual fillers form multi-layer aggregation caused by incompatibility with polymer matrix. One consequence of the described phenomenon is that the water-vapor transmission rate (WVTR) varies considerably in the layer leading to unsatisfying average values for the WVTR. Secondly, the low dispersion of the filler causes an

easy upcurling of the polymer composition coated on the materials. Hence, a uniform dispersion of fillers incorporated in a polymer composition should improve the water-vapor transmission rate significantly, and, additionally, the curling properties of a coated material should be enhanced.

Hence, the object of the present invention is to improve the water-vapor transmission rate (WVTR).

The present invention is based on the finding that the object can be addressed by a polymer composition comprising a polymer having a low average molecular weight enabling an enhanced and uniform dispersion of fillers incorporated in the polymer composition.

The present invention therefore provides a multimodal polymer composition comprising

- a) at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60,000 g/mol;
- b) at least one polyolefin (B) having a higher weight average molecular weight (M_w) than polymer (A); and
- c) a filler (C) whereby the polymer composition without filler (C) has a density of 940 kg/m³ or lower.

It is preferred that the polymer composition consists of

- a) at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60,000 g/mol;
- b) at least one polyolefin (B) having a higher weight average molecular weight (M_w) than polymer (A); and
- c) a filler (C) whereby

the polymer composition without filler (C) has a density of 940 kg/m³ or lower.

Accordingly the polymer composition according to this invention is multimodal with respect to the molecular weight distribution. "Multi-modal" or "multimodal distribution" describes a frequency distribution that has several relative maxima. In particular, the expression "modality of a polymer" refers to the form of its molecular weight distribution (MWD) curve, i.e. the appearance of the graph of the polymer weight fraction as a function of its molecular weight. The molecular weight distribution (MWD) of a polymer produced in a single polymerization stage using a single monomer mixture, a single polymerization catalyst and a single set of process conditions (i.e. temperature, pressure, etc.) shows a single maximum the breadth of which depends on catalyst choice, reactor choice, process conditions, etc., i.e. such a polymer is monomodal.

This inventive composition is characterized by a very low water-vapor transmission rate (WVTR) and also by low curling-values for extrusion-coated layers. These improved properties are reached by a much better dispersion of the filler (C) in the polymer mixture of polymer (A) and polyolefin (B) compared with an unimodal polymer having the same melt index and density for both extrusion-coated layers and cast films.

Hence, the polymer composition according to this invention is a multimodal including bimodal polymer composition consisting of at least two different polymers having two different molecular weight distribution curves and are blended mechanically or in situ during the preparation thereof. Preferably the polymer composition is at least a bimodal mechanical or in-situ blend of a polyolefin (1) (as polymer (A)) and polymer (B). In case such a bimodal blend comprises further a wax (2) as an additional polymer (A), then the final polymer composition may also be trimodal.

The molecular weight distribution (MWD) is the relation between the numbers or molecules in a polymer and their individual chain length. The molecular weight distribution (MWD) is often given as a number, which normally means weight average molecular weight (M_w) divided by number average molecular weight (M_n).

The weight average molecular weight (M_w) is the first moment of a plot of the weight of polymers in each molecular weight range against molecular weight. In turn, the number average molecular weight (M_n) is an average molecular weight of a polymer expressed as the first moment of a plot of the number of molecules in each molecular weight range against the molecular weight. In effect, this is the total molecular weight of all molecules divided by the number of molecules.

The number average molecular weight (M_n) and the weight average molecular weight (M_w) as well as the molecular weight distribution (MWD) are determined according to ISO 16014.

The weight average molecular weight (M_w) is a parameter for the length of the molecules in average. Low M_w -values indicate that the chain length of the molecules is rather short in average. It has been found out that a polymer mixture comprising a polymer (A) with M_w -values of lower than 60,000 g/mol contributes inter alia to better barrier properties and better dispersion of the filler (C). Such better dispersion improves the water-vapor transmission rate (WVTR) as well as the curling resistance positively.

Hence, as a further requirement of the present invention, the multimodal polymer composition must comprise at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60,000 g/mol. It is in particular preferred that at least one polymer (A) having a weight average molecular weight (M_w) of

lower than 60,000 g/mol is at least one polyolefin (1) having a weight average molecular weight (M_w) of 10,000 to 60,000 g/mol, more preferably of 20,000 to 50,000 g/mol and/or at least one wax (2) having a weight average molecular weight (M_w) of less than 10,000 g/mol, more preferably in the range of 500 to 10,000 g/mol.

Moreover, it is preferred that the polyolefin (1) is a polyethylene or polypropylene, more preferably a polyethylene. The polyolefin (1) can be a homopolymer or copolymer. It is preferred that the polyolefin (1) is a homopolymer or copolymer of propylene or ethylene, more preferred the polyolefin (1) is a homopolymer or copolymer of ethylene. Most preferably the polyolefin (1) is a low density polyethylene (LDPE), a linear low density polyethylene (LLDPE), or a linear medium density polyethylene (LMDPE). LDPE, LLDPE and LMDPE are equally suitable alternatives for polyolefin (1), e.g. where a LLDPE or a LMDPE is applicable also a LDPE can be used and vice versa.

In case polymer (A) is a wax (2), it is preferred that it is selected from one or more of

- (2a) a polypropylene wax having a weight average molecular weight (M_w) of less than 10,000 g/mol, more preferably in the range of 500 to 10,000 g/mol, still more preferably in the range of 1000 to 9000 g/mol, yet more preferably in the range of 2000 to 8000 g/mol and most preferably in the range of 4000 to 8000 g/mol or a polyethylene wax having a weight average molecular weight (M_w) of less than 10,000 g/mol, more preferably in the range of 500 to 10,000 g/mol, still more preferably in the range of 1000 to 9000 g/mol, yet more preferably in the range of 2000 to 8000 g/mol and most preferably in the range of 4000 to 8000 g/mol, and
- (2b) an alkyl ketene dimer wax having weight average molecular weight (M_w) of less than 10,000 g/mol, more preferably lower than 5000 g/mol, yet

more preferably lower than 1000 g/mol. In turn the alkyl ketene dimer wax has preferably weight average molecular weight (M_w) of at least 100 g/mol. Most preferred the alkyl ketene dimer wax has weight average molecular weight (M_w) in the range of 250 to 1000 g/mol.

The terms "at least one polymer (A)", "at least one polyolefin (1)" or "at least one wax (2)" shall indicate that more than one polymer (A), polyolefin (1) or wax (2) can be present in the multimodal polymer composition. It is preferred that three, two or one different polymers (A) as defined above are used in a multimodal polymer composition. Still more preferred is that wax (2), preferably a polypropylene wax (2a) or an alkyl ketene dimer wax (2b) as defined above is used as a component (A) only. In case the component (A) comprises a polyolefin (1) as defined above, it is preferred that a wax (2) is present in the multimodal polymer composition as a further polymer (A). In such cases the multimodal composition is preferably trimodal comprising polyolefin (1), wax (2) and polyolefin (B) having different centered maxima in their molecular weight distribution, e.g. having different weight average molecular weights (M_w). The use of the wax (2) has the benefit that the amorphous region of the polymer matrix, which may be a mix of polyolefin (1) and polyolefin (B), is filled up and improves thereby the barrier properties.

It is preferred that not only the final polymer composition has a specific density of 940 kg/m³ or lower but also the polymer (A) shall have a density of lower than 945 kg/m³. It is preferred that polyolefin (1) when used as polymer (A) has a density lower than 945 kg/m³, more preferably is in a range of 905 to 935 kg/m³, still more preferably in the range of 910 to 930 kg/m³ and most preferred in the range of 915 to 925 kg/m³. Preferably, the polyolefin (1) is a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE). In turn, also a low density polyethylene (LDPE) produced in a high pressure process by free radical polymerization is applicable as a polyolefin (1). The linear low

density polyethylene (LLDPE) or the linear medium density polyethylene (LMDPE) is produced by a process as described for the polyolefin (B) below.

The molecular weight distribution (MWD) of the polymer composition is further characterized by the way of its melt flow rate (MFR) according to ISO 1133 at 190°C. The melt flow rate (MFR) mainly depends on the average molecular weight. The reason for this is that long molecules give the material a lower flow tendency than short molecules.

An increase in molecular weight means a decrease in the MFR-value. The melt flow rate (MFR) is measured in g/10 min of the polymer discharged under specific temperature and pressure conditions and is the measure of a viscosity of the polymer which in turn for each type of polymer is mainly influenced by its molecular weight distribution, but also by its degree of branching. The melt flow rate measured under a load of 2.16 kg (ISO 1133) is denoted as MFR₂. In turn, the melt flow rate measured with 5 kg load (ISO 1133) is denoted as MFR₅.

In case polymer (A) is a polyolefin (1), it is preferred that MFR₂ is in the range of 1.0 to 20.0 g/10 min and more preferably in the range of 2.0 to 15.0 g/10 min and for example in the range of 3.0 to 10.0 g/10 min. According to one embodiment, the polyolefin (1) is a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE) with MFR₂ as given above. In turn, polyolefin (1) can also be a low density polyethylene (LDPE) having a density in the range as stated in this paragraph. The low density polyethylene (LDPE) is produced in a high-pressure process by free radical polymerization. In turn, the linear low density polyethylene (LLDPE) or the linear medium density polyethylene (LMDPE) is produced as described for polyolefin (B).

In case polymer (A) is an ethylene homopolymer, it is preferred that the ethylene homopolymer contains less than 0.2 mol%, more preferably less than 0.1 mol%

and most preferably less than 0.05 mol% units derived from alpha-olefins other than ethylene. It is in particular preferred that the polymer (A) is an ethylene copolymer, more preferably having a weight average molecular weight from 10,000 to 60,000 g/mol, still more preferably from 20,000 to 50,000 g/mol. Yet more preferably the polymer (A) is an ethylene copolymer having a density of 905 to 935 kg/m³, more preferably of 910 to 930 kg/m³, most preferably of 915 to 925 kg/m³ and having a melt index MFR₂ from 1.0 to 20.0 g/10 min, more preferably from 2.0 to 15.0 g/10 min, most preferably of 3.0 to 10.0 g/10 min. Preferably, the ethylene copolymer comprises, more preferably consists of, comonomer units as defined below for the LLDPE. It is in particular preferred that the ethylene copolymer fulfills all properties as stated above simultaneously.

In case polymer (A) is a wax (2a), namely a polypropylene wax or a polyethylene wax, it is preferred that the wax (2a) has a weight average molecular weight (M_w) in the range of 500 to 10,000 g/mol, more preferably in the range of 1,000 to 9,000 g/mol, still more preferably in the range of 2,000 to 8,000 g/mol and most preferably in the range of 4,000 to 8,000 g/mol. Further preferred ranges for the weight average molecular weight (M_w) of the wax (2a), in particular the polypropylene or polyethylene wax, is in the range of 4,000 to 7,000 g/mol, still more preferably in the range of 5,000 to 6,000 g/mol and most preferably in the range of 5,300 to 5,400 g/mol. Additionally, it is preferred that the wax (2a), in particular the polypropylene wax or polyethylene wax, has a z-average molecular weight of 9,100 to 40,000 g/mol, more preferably from 500 to 20,000 g/mol and most preferably from 10,000 to 12,000 g/mol. It is additionally preferred that the wax (2a), in particular the polypropylene wax or the polyethylene wax, has a number average molecular weight (M_n) of 100 to 20,000 g/mol, more preferably of 500 to 3,000 g/mol.

Moreover, it is preferred that wax (2a), in particular polypropylene wax or polyethylene wax, has a specific molecular weight distribution (MWD) which is the

relation between the number of molecules in the polymer and their individual chain length. The molecular weight distribution is given as a number which means weight average molecular weight divided by number average molecular weight (M_w/M_n). It is preferred that the wax (2a), in particular the polypropylene wax or the polyethylene wax, has an MWD in the range of 1 to 5, more preferably in the range of 1.5 to 4.

In addition, it is preferred that the wax (2a), in particular the polypropylene wax or the polyethylene wax, has a melting temperature in DSC-analysis of below 150°C, more preferably below 140°C, still more preferably in the range of 95 to 130°C, most preferably in a range of 105 to 115°C.

In case a wax (2b), namely an alkyl-ketene dimer, is employed as polymer (A), it is preferred that the weight average molecular weight (M_w) of the wax (2b) is higher than 100 g/mol. In turn, it is preferred that the weight average molecular weight of the wax (2b) is lower than 10,000 g/mol, more preferably lower than 5,000 g/mol, still more preferably lower than 1,000 g/mol. Preferred ranges for the weight average molecular weight (M_w) of the wax (2b) is 100 to 10,000 g/mol, more preferably 250 to 1,000 g/mol. Additionally, it is preferred that the wax (2b) has a number average molecular weight (M_n) of 100 to 20,000 g/mol, more preferably in the range of 100 to 800 g/mol. In addition, it is preferred that wax (2b) has a melting temperature in DSC-analysis below 140°C, more preferably below 100°C. A preferred range for the melting temperature in DSC-analysis is 50 to 90°C, more preferably 50 to 70°C.

As a further requirement, according to the present invention, the polyolefin (B) shall have a higher weight average molecular weight (M_w) than polymer (A). It is preferred that the polyolefin (B) has a weight average molecular weight (M_w) of higher than 80,000 g/mol, more preferably higher than 100,000 g/mol. The upper limit for the weight average molecular weight for polyolefin (B) shall preferably

not be higher than 300,000 g/mol, more preferably not higher than 200,000 g/mol. The preferred range for the weight average molecular weight for polyolefin (B) is 80,000 to 300,000 g/mol, more preferably from 100,000 to 200,000 g/mol. Preferably, polyolefin (B) is a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE), which has been preferably produced in a low medium pressure process in the presence of a polymerization catalyst (i.e. a Ziegler-Natta catalyst or a metallocene catalyst). A linear low density polyethylene (LLDPE) and a linear medium density polyethylene (LMDPE) has a density lower than 945 kg/m³, more preferably in the range of 905 to 935 kg/m³, still more preferably in the range of 910 to 930 kg/m³ and most preferred in the range of 915 to 925 kg/m³. However, also a low density polyethylene (LDPE) is also applicable for the polyolefin (B). A low density polyethylene (LDPE) has the same density ranges as the LLDPE or the LMDPE as stated in this paragraph and is a product from a high pressure polymerization process characterized by a highly branched chain structure. LDPE, LLDPE and LMDPE are equally suitable alternatives for polyolefin (B), e.g. where a LLDPE or a LMDPE is applicable also a LDPE can be used and vice versa.

According to this invention, more than one polyolefin (B) can be used. Accordingly, the invention also comprises the possibility of any mixture of a linear low density polyethylene (LLDPE), a linear medium density polyethylene (LMDPE) and a low density polyethylene (LDPE).

The MFR₂ of the polyolefin (B) is preferably in the range of 1.0 to 20.0 g/10 min, more preferably in the range of 2.0 to 15.0 g/10 min, in the range of e.g. 3.0 to 10.0 g/10 min. It is in particular preferred that the linear low density polyethylene (LLDPE) and the linear medium density polyethylene (LMDPE) have such melt flow characteristics. In turn, also the low density polyethylene (LDPE) suitable as a polyolefin (B) may have the melt flow characteristics as given in this paragraph.

It is preferred that the polyolefin (B) is a polyethylene. In case the polyolefin (B) is a polyethylene, it may be an ethylene homopolymer or an ethylene copolymer. In case for the polyolefin (B) an ethylene homopolymer is employed, then preferably an ethylene homopolymer is used as defined for polymer (A). In case an ethylene copolymer is employed for polyolefin (B), then preferably an ethylene copolymer is used as defined below. It is in particular preferred that polyolefin (B) is a low density polyethylene (LDPE), a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE).

According to one embodiment, the polymer composition according to this invention is a linear low density polyethylene (LLDPE) comprising polyolefin (1) (polymer (A)) as a low molecular weight fraction of LLDPE and polyolefin (B) as a high molecular weight fraction of LLDPE. This linear low density polyethylene (LLDPE) may be a mechanical blend, preferably an in-situ blend produced in a multistage process. Preferably said composition comprises wax (2) as a further polymer (A).

It is preferred that the polymer composition as defined above comprises 1 to 50 wt% of polymer (A), 40 to 90 wt% of polyolefin (B) and 1 to 50 wt% of filler (C), more preferably of 5 to 40 wt%, and most preferably of 10 to 35 wt%. In case the polymer composition is produced in an in situ polymerization process, e.g. a sequential step process by utilizing reactors coupled in series and described as above, it is preferred that the polymer (A) may range from 40 to 60 wt%, more preferably 49 to 55 wt% in the polymer mix without filler (C). In turn, it is preferred that in such a polymer mix, the polyolefin (B) ranges from 60 to 40 wt%, more preferably from 51 to 45 wt%. Preferably, the total polymer composition comprises 50 to 99 wt% of said polymer mix and of 1 to 50 wt% filler (C), more preferably of 5 to 40 wt%, and most preferably of 10 to 35 wt%.

In case polymer (A) and polyolefin (B) are blended mechanically, it is preferred that polymer (A) ranges from 1 to 30 wt% and, more preferably, from 1 to 20 wt% in the total polymer composition. These ranges apply in particular in case for polymer (A) a wax (2) is used only.

The last requirement according to the present invention is that the multimodal polymer composition additionally comprises a filler (C). Any filler having a positive influence on the water-vapor transmission rate (WVTR) can be used. Preferably, the filler shall be lamellar, such as clay, mica or talc. More preferably, the filler shall be finely divided. The finely divided filler consists of about 95 wt% of particles having particle sizes of less than 10 μm , and about 20-30 wt% of particles having a particle size of less than 1 μm . In the present invention all layer materials may be used as long as they have the ability to disperse in the polymer composition. The filler may either be a clay-based compound or a submicron filler such as talc, calcium carbonate or mica, which usually have been treated, for instance by grinding, to obtain particles of small, i.e. submicron, dimensions, in situ as stated above.

It is preferred that the filler (C) is layered silicate material, still more preferred, filler (C) is a clay-based compound. Clay-based compounds upon compounding of the polymer composition are dispersed in the polymer composition so that individual platelets in the layered structure are separated.

In a further preferred embodiment, the filler (C) is a clay-based layered inorganic, preferably silicate material or material mixture. Such useful clay materials include natural, synthetic and modified phyllosilicates. Natural clays include smectite clays, such as montmorillonite, hectorite, mica, vermiculate, bentonite. Synthetic clays include synthetic mica, synthetic saponite, synthetic hectorite. Modified clays include fluorinated montmorillonite, fluorinated mica.

Of course, the filler (C) may also contain components comprising a mixture of different fillers, such as mixtures of a clay-based filler and talc.

Layered silicates may be made organophylic before being dispersed in the polymer composition by chemical modification, such as by cation-exchange treatment using alkyl ammonium or phosphonium cation complexes. Such cation complexes intercalate between the clay layers.

Preferably, a smectite type clay is used, which comprises montmorinollite, beidellite, nontronite, saponite, as well as hectonite. The most preferred smectite type clay is montmorinollite.

Preferably, also talc is used as a filler (C).

The density affects most physical properties like stiffness impact strength and optical properties of the end products. Hence, and according to the present invention, the density of the polymer composition shall be of 945 kg/m^3 or lower. More preferably, the density shall range from 905 to 935 kg/m^3 , still more preferably from 910 to 930 kg/m^3 and most preferably from 915 to 925 kg/m^3 .

The ranges and values given for the density in the whole invention apply for pure polymer compositions and do not include any additives, in particular no filler (C). The density is determined according to ISO 1183-1987.

Moreover, it is preferred that the polymer composition without any additive, preferably without filler (C) has a melt flow rate MFR_2 according to ISO 1133 at 190°C of 5 to 20 g/10 min , more preferably from 7 to 15 g/10 min .

Preferably, the polymer composition without any additive, preferably without filler (C) has a melt flow rate MFR_5 according to ISO 1133 at 190°C of 20 to 40 g/10 min, more preferably of 25 to 35 g/10 min.

Moreover, it is preferred that the melt flow ratio, which is a ratio of two melt flow rates measured for the same polymer under two different loads, falls within a specific range. The preferred specific range is 2.5 to 4.5, more preferably 2.7 to 4.0, for the melt flow ratio MFR_5/MFR_2 .

A further characteristic of the molecular weight distribution (MWD) which is the relation between the number of molecules in a polymer and their individual chain length has to be considered. The width of the distribution is a number as a result of the ratio of the weight average molecular weight divided by the number average molecular weight (M_w/M_n). In the present invention, it is preferred that the polymer composition without any additive, preferably without filler (C), has a M_w/M_n of preferably 8 to 25 and more preferably from 10 to 20.

Additional additives, e.g. inorganic additives, known as expipients and extrusion aids in the field of coatings and films, are used.

For a better adhesion between the coating and the substrate, it is preferred that the polymer is oxidized. Consequently, it is preferred that the polymer composition contains anti-oxidants and process stabilizers less than 2,000 ppm, more preferably less than 1,000 ppm and most preferably not more than 700 ppm. The anti-oxidants thereby may be selected from those known in the art like those containing hindered phenols, secondary aromatic amines, thio-ethers or other sulfur-containing compounds, phosphites and the like including their mixtures.

It has been found that the polymer composition as described above has a very low water-vapor transmission rate (WVTR). Additionally, the composition has a

good adhesion to the substrate, in particular to aluminium, without any need to have an adhesion layer between the substrate and the coating. Further, the tendency of the coated article to curl is significantly reduced for the polymer composition compared to neat polymer. These advantageous effects could only be achieved as the miscibility between the polymer and the filler is much higher for a multimodal or bimodal polymer having a low molecular weight polymer fraction in comparison with a polymer having the same melt index and density.

In one preferable embodiment, the multimodal composition comprises as polymer (A), which is the low molecular weight fraction, a polyolefin (1), more preferably a low density polyethylene (LDPE) or linear low density polyethylene (LLDPE). The polyolefin (B), which is the high molecular weight fraction, is a low density polyethylene (LDPE) or a linear low density polyethylene (LLDPE). Preferably, this composition comprises a further polymer (A) which is a wax (2) as defined above. This composition can be produced in an in situ process or can be blended mechanically. Preferred properties for the polymer (A), in particular the polyolefin (1), the wax (2) and the polyolefin (B) are those as given above. In case this composition comprises two polymers (A), namely a polyolefin (1) and a wax (2), it is preferred that the amount of wax (2) in the total composition without filler (C) is 1 to 30 wt%, more preferably 1 to 20 wt% and most preferably 1 to 10 wt%. In turn, the composition comprises 70 to 99 wt%, more preferably 80 to 99 wt% and most preferably 90 to 99 wt% of LLDPE resulting from polymer (A) and polyolefin (B). In case the composition comprises LDPE, it is preferred that wax (2) is present in the amount of 1 to 30 wt% and LDPE resulting at least from polymer (B) and optionally from polymer (A) and is present in the amount of 70 to 99 wt% in the total composition without filler (C).

In another preferable embodiment, a polymer composition is produced in an in situ process, whereby the sequential step process by utilizing reactors coupled in series as described above is preferred. Preferably polymer (A) is produced in a

loop reactor whereas polyolefin (B) is produced in a gas phase reactor in the presence of polymer (A). Thereby, it is preferred that the multimodal polymer is at least a bimodal polymer. More preferably, polymer (A) and polyolefin (B) are both polyolefins. The polymer composition of this embodiment comprises 50 to 99 wt% of a linear low density polyethylene (LLDPE) having a multimodal, more preferably a bimodal molecular weight distribution (MWD) and more preferably 1 to 50 wt% of a filler (C), preferably a plate- or sheet-like filler such as mica or talc as described above.

In the following, when the description refers to LLDPE, it means that a multimodal, preferably bimodal LLDPE is used, which comprises a low molecular weight (LMW) fraction, which is polymer (A) (polyolefin (1)), and a high molecular weight (HMW) fraction, which is polymer (B).

Preferably, the linear low density polyethylene (LLDPE) has a melt index MFR_2 from 1.0 to 20 g/10 min, more preferably from 2 to 15 g/10 min and most preferably from 3 to 10 g/10min. It is preferred that the linear low density polyethylene (LLDPE) is at below 945 and ranges preferably from 905 to 935 kg/m³, more preferably from 910 to 930 kg/m³, most preferably from 915 to 925 kg/m³. If the melt index of the linear low density polyethylene (LLDPE) is lower than 1 g/10 min, a high throughput is not reached. On the other hand, if the melt index MFR_2 is higher than 20, the melt strength of the polyethylene suffers.

In addition, it is preferred that the linear low density polyethylene (LLDPE) has a melt flow index MFR_5 from 20 to 40 and preferably a melt flow ratio MFR_5/MFR_2 from 2.5 to 4.5; more preferably from 2.7 to 4.0. Furthermore, it is preferred that the linear low density polyethylene (LLDPE) has a weight average molecular weight (M_w) from 50,000 to 150,000 g/mol, more preferably from 60,000 to 100,000 g/mol and preferably a ratio of the weight average molecular

weight to the number average molecular weight M_w/M_n of 8 to 25, more preferably of 10 to 20.

Moreover, the linear low density polyethylene (LLDPE) contains comonomers selected from the group consisting of C_3 alpha-olefin, C_4 alpha-olefin, C_5 alpha-olefin, C_6 alpha-olefin, C_7 alpha-olefin, C_8 alpha-olefin, C_9 alpha-olefin, C_{10} alpha-olefin, C_{11} alpha-olefin, C_{12} alpha-olefin, C_{13} alpha-olefin, C_{14} alpha-olefin, C_{15} alpha-olefin, C_{16} alpha-olefin, C_{17} alpha-olefin, C_{18} alpha-olefin, C_{19} alpha-olefin, C_{20} alpha-olefin. Especially preferred are alpha-olefins selected from the group consisting of propylene, 1-butene, 1-pentene, 4-methyl-1-pentene, 1-hexene, 1-heptene, 1-octene, 1-nonene, 1-decene, 6-methyl-1-heptene, 4-ethyl-1-hexene, 6-ethyl-1-octene and 7-methyl-1-octene. Still more preferred, alpha-olefins are selected from the group consisting of 1-butene, 4-methyl-1-pentene, 1-hexene and 1-octene.

As one requirement of the preferred embodiment is that the polymer composition is a linear low density polyethylene (LLDPE) the content of the comonomer units in the polymer is preferably 0.1 to 1.0 mol%, more preferably 0.15 to 0.5 mol%.

It is preferred that the linear low density polyethylene (LLDPE) without filler (C) comprises 40 to 60 wt%, more preferably 49 to 55 wt% polymer (A) and 60 to 40 wt%, and more preferably 51 to 45 wt% polyolefin (B).

As stated above, it is preferred that the linear low density polyethylene (LLDPE) comprises a LMW fraction, which forms the polymer (A). More preferably, the polymer (A), is a polyolefin (1), most preferably an ethylene copolymer containing alpha-olefins other than ethylene and listed above. Furthermore, it is preferred that the polymer (A) of the linear low density polyethylene (LLDPE) has a weight average molecular weight (M_w) of 10,000 to 60,000 g/mol, more preferably from 20,000 to 50,000 g/mol. It is further preferred that polymer (A) of the

linear high density polyethylene (LLDPE), has a density of at least 905 to 935 kg/m³, more preferably of at least 910 to 930 kg/m³. In addition, it is preferred that polymer (A) of the linear low density polyethylene (LLDPE) has a melt flow rate MFR₂ from 1.0 to 20.0 g/10 min, more preferably from 2.0 to 15.0 g/10 min and most preferred from 3 to 10 g/10min.

It is preferred that polyolefin (B) as the linear low density polyethylene (LLDPE) is an ethylene copolymer containing one or more alpha-olefins as listed above. Thereby, it is preferred that the amount of comonomer units in polyolefin (B) is from 2.0 to 15.0mol%, more preferably from 3.0 to 10.0 mol%. In addition, it is preferred that the polyolefin (B) in the linear low density polyethylene (LLDPE) has a weight average molecular weight from 80,000 to 300,000 g/mol, more preferably from 100,000 to 200,000 g/mol.

The filler (C) and other additional components in the linear low density polyethylene (LLDPE), are identically used as listed and described above. It is in particular preferred that additionally to the LLDPE, a wax (2), more preferably a polypropylene wax (2a) or an alkyl-ketene dimer (2b) as defined above is used as an additional polymer (A).

In case two polymers (A) are used, namely polyolefin (1) and wax (2), the amount of wax (2) is 1 to 30 wt%, more preferably 2 to 20 wt% and most preferably 1 to 10 wt% in the total composition without filler (C). In turn, the composition without filler (C) comprises 70 to 99 wt%, more preferably 80 to 88 wt% and most preferably 90 to 99 wt% LLDPE resulting from polymer (A) and polyolefin (B).

The further preferable embodiment of the present invention is a polymer composition whereby polymer (A) and polyolefin (B) are preferably mechanically

blended. Thereby it is preferred that polymer (A) is a wax (2), more preferably a polypropylene wax (2a) or an alkyl-ketene dimer wax (2b).

In case of polymer (A), where a polypropylene wax (2a) is used, it is preferred that this wax (2a) has a weight average molecular weight (M_w) of 100 to 50,000, more preferably from 100 to 10,000, and most preferably from 5,000 to 6,000. In addition, it is preferred that the z-average molecular weight of the polypropylene wax (2a) ranges from 100 to 60,000 g/mol, and more preferably from 100 to 10,000 g/mol. It is preferred that the polypropylene wax (2a) has a number average molecular weight (M_n) of 100 to 2,000 g/mol, more preferably 500 to 3,000 g/mol. The melting temperature in DSC-analysis of the polypropylene wax (2a) is preferably of 95 to 130°C, more preferably 105 to 115°C.

Preferably, the polypropylene wax (2a) is mechanically blended with an ethylene polymer as a polyolefin (B) having an MFR_2 of 6.5 to 8.5 g/10 min, more preferably from 7 to 8 g/10 min and a density of 900 to 940 kg/m³, more preferably from 915 to 925 kg/m³. It is in particular preferred that polyolefin (B) is a low density polyethylene (LDPE), or a linear low density polyethylene (LLDPE) as described above.

The mechanically blended polymer including a talc as filler (C) and CaO as a water-absorbent component has preferably a density ranging from 1,000 kg/m³ to 1,300 kg/m³, more preferably of 1,150 to 1,200 kg/m³ and a melt flow rate MFR_2 of preferably 8 to 9.5 g/10 min, and more preferably of 8.5 to 9.0 g/10 min.

The other preferred alternative of a mechanical blend of wax (2) with polyolefin (B) is to use an alkyl-ketene dimer (2b) as wax (2). Preferably, this alkyl-ketene dimer (2b) has a weight average molecular weight (M_w) of 300 to 400 g/mol, more preferably from 320 to 350 g/mol. Preferably, the z-average molecular weight of the alkyl-ketene dimer (2b) is from 300 to 400 g/mol, more preferably

from 360 to 390 g/mol. It is preferred that the alkyl-ketene dimer (2b) has a number average molecular weight (M_n) of 200 to 450 g/mol, more preferably from 280 to 300 g/mol. In addition, it is preferred that the alkyl-ketene dimer (2b) has a melting temperature DSC-analysis of 55 to 70°C, more preferably from 60 to 65°C.

For polyolefin (B), the same ethylene polymer is used as defined under the mechanical blend comprising a polypropylene wax (2a).

The density of the mechanically blended polymer composition comprising an alkyl-ketene dimer (2b) as defined above, an ethylene polymer (B) as defined above, a filler (C) and a water-absorbent component has preferably a density of 1,050 to 1,300 kg/m³ and more preferably from 1,150 to 1,250 kg/m³. The melt flow rate MFR₂ of this polymer composition is preferably from 12.5 g/10 min to 14.5 g/10 min and more preferably from 13 to 14 g/10 min. It is preferred that for this embodiment for filler (C) talc is employed and for the water absorbent compound CaO.

Furthermore, the present invention comprises a process for producing the multimodal composition as defined above.

A multimodal or at least bimodal, e.g. bimodal or trimodal, polymer may be produced by blending two or more monomodal polymers having differently centered maxima in their molecular weight distributions. The blending may be effected mechanically, e.g. analogously to the mechanical blending principles known in the art. Alternatively, the multimodal or at least bimodal, e.g. bimodal or trimodal, polymer composition may be produced by polymerization using conditions which create a multimodal or at least bimodal, e.g. bimodal or trimodal, polymer composition, i.e. using a catalyst system for mixtures with two or more different catalytic sites, using two or more stage polymerization process with different

process conditions in the different stages (i.e. different temperatures, pressures, polymerization media, hydrogen partial pressures, etc.). With the polymer as produced in such a sequential step process, i.e. by utilizing reactors coupled in series, and using different conditions in each reactor, the different polymer fractions produced in the different reactors will each have their own molecular weight distribution which may differ considerably from one another. The molecular weight distribution curve of the resulting final polymer can be regarded as superimposing of the molecular weight distribution curves of the polymer fractions which will accordingly show two or more distinct maxima, or at least the distinctively broadened maxima compared with the curves for individual fractions.

A polymer showing such a molecular weight distribution curve is called multimodal, trimodal or bimodal.

Multimodal polymers can be produced according to several processes, which are described, e.g. in WO 92/12182 and WO 97/22633.

A multimodal polymer is preferably produced in a multi-stage process in a multi-stage reaction sequence, such as described in WO 92/12182. The contents of this document are included herein by reference.

It is known to produce multimodal or at least bimodal, e.g. bimodal or trimodal, polymers, preferably multimodal or bimodal olefin-polymers, such as multimodal or bimodal polyethylenes in two or more reactors connected in series whereby the compounds (A) and (B) can be produced in any order.

According to the present invention, the main polymerization stages are preferably carried out as a combination of a slurry gas/gas-phase polymerization. The slurry polymerization is preferably performed in a so-called loop-reactor.

Optionally, and of more advantage, the main polymerization stages may be preceded by a pre-polymerization in which case up to 20 wt%, preferably 1-10 wt%, more preferably 1-5 wt% of the total amount of polymer composition is produced. At the pre-polymerization point, all of the catalyst is preferably charged into a loop-reactor and a polymerization is performed as a slurry polymerization. Such a polymerization leads to less fine particles being produced in the following reactors and to a more homogeneous product being obtained in the end. Such a pre-polymerization is for instance described in WO 96/18662.

Generally, the technique results in a multimodal or at least bimodal, e.g. bimodal or trimodal, polymer composition thereby a Ziegler-Natta or metallocene catalyst in several successive polymerization reactors is used. For example in the production of a bimodal high-density polyethylene composition, a first ethylene polymer is produced in the first reactor under certain conditions with respect to the hydrogen-gas concentration, temperature, pressure and so forth. After the polymerization the reactor-polymer including the catalyst is separated from the reaction mixture and transferred to a second reactor where further polymerization takes place under other conditions.

The components (A) and (B) can be produced with any suitable catalyst system, preferably a coordination catalyst, such as a Ziegler-Natta catalyst system, preferably a coordination catalyst, such as a Ziegler-Natta catalyst of a transition metal of a group 3-10 of the periodic table (IUPAC), a metallocene, non-metallocene, in a manner known in the art. One example of a preferred Ziegler-Natta catalyst comprises Ti, Mg and Al, such as described in document EP 0 688 794 B1, which is included herewith by reference. It is a high-activity pro-catalyst comprising a particular inorganic support, a curing compound deposited on the support, wherein the curing compound is the same as or different from the titanium compound, whereby the inorganic support is contacted with an alkyl metal

chloride which is soluble in a non-polar hydrocarbon solvent, and has the formula $(R_nMeCl_{3-n})_m$, wherein R is a C_1 to C_{20} alkyl group, Me is a metal of Group III(13) of the periodic table, $n = 1$ or 2 and $m = 1$ or 2 , to give a first reaction product, and the first reaction product is contacted with a compound containing hydrocarbyl and hydrocarbyl oxide linked to magnesium which is soluble in non-polar hydrocarbon solvents, to give a second reaction product, and the second reaction product is contacted with a titanium compound which contains chlorine, having the formula $Cl_xTi(OR^{IV})_{4-x}$, wherein R^{IV} is a C_2 to C_{20} hydrocarbyl group and $x = 3$ or 4 , to give the procatalyst. Preferred supports are inorganic oxides, more preferably silicon dioxide or silica. Most preferably silica having an average particle size of $20\ \mu m$ is used. Even more preferred tri-ethyl aluminium as a cocatalyst is used. Alternatively, a metallocene of group 4 metal can be used.

Preferably, polymer (A), the low molecular weight (LMW) polymer, is produced with addition or no addition of comonomer in a first reactor, and also the polyolefin (B), the high molecular weight (HMW) polymer, is produced with addition or no addition, more preferably with addition, of comonomer in the second reactor.

The resulting end product consists of an intimate mixture of polymers from the two reactors, the different molecular weight distribution occurs of these polymers together forming a molecular weight distribution curve having a broad maximum or two maxima, i.e. the end product is a multimodal or bimodal polymer mixture. Since multimodal and, in particular, bimodal polymers, preferably ethylene polymers and the production thereof belong to the prior art, no detailed description is called for here, but reference is made to the above-mentioned document WO 92/12182. It will be noted that the order of the reaction stages may be reversed.

Preferably, as stated above, the multimodal polymer composition according to the invention is a bimodal or trimodal polymer composition. It is also preferred that this bimodal or trimodal polymer composition has been produced by polymerization as described above under different polymerization conditions in two or more polymerization reactors connected in series.

Furthermore, it is preferred that for the multimodal composition according to this invention a process is used as defined above whereby

- a) polymer (A) and polyolefin (B) are produced together in a multi-stage process comprising a loop reactor and a gas-phase reactor, wherein polymer (A) is generated in at least one loop reactor and the polyolefin (B) is generated in a gas-phase reactor in the presence of the reaction product (A) of the loop reactor, and
- b) filler (C) and the composition comprising polymer (A) and polyolefin (B) are blended together and compounded.

In particular, a multi-stage process is used as described above. Especially, it is preferred that a loop reactor is operated at 75 to 100°C, more preferably in the range of 85 to 100°C and most preferably in the range of 90 to 98°C. Thereby, the pressure is preferably 58 to 68 bar, more preferably 60 to 65 bar.

Preferably, polymer (A) is prepolymerized in a first loop reactor and then continuously removed to a second loop reactor where the polymer (A) is further polymerized. It is preferred that the temperature in the second loop reactor is 90 to 98°C, more preferably about 95°C. Thereby, the pressure is preferably 58 to 68 bar, more preferably about 60 bar.

In addition, it is preferred that in the second loop reactor, the ethylene concentration is 4 to 10 mol%, more preferably 5 to 8 mol% and most preferably about 6.7 mol%.

The hydrogen to ethylene mol-ratio highly depends on the catalyst used. It must be adjusted to render the desired melt flow rate MFR of the polymer withdrawn from the loop reactor. For the preferred catalyst as described it is preferred that the ratio of hydrogen to ethylene is 100 to 800 mol/kmol and more preferably 300 to 700 mol/kmol, still more preferably 400 to 650 mol/kmol and most preferred about 550 mol/kmol.

The polymer slurry is then preferably removed from the loop reactor by using settling tanks and is then preferably introduced into a flash vessel operating preferably at about 3 bar pressure, where the polymer is separated from most of the fluid phase. The polymer is then preferably transferred into a gas-phase reactor operating preferably at 75 to 95°C, more preferably 80 to 90°C and most preferably about 85°C, and at preferably 10 to 50 bar, more preferably 15 to 25 bar and most preferably about 20 bar.

Additionally, ethylene comonomers were used and hydrogen as well as nitrogen as an inert gas are preferably introduced into the reactor so that the fractional ethylene in the fluidization gas is preferably 1 to 10 mol%, more preferably 1 to 5 mol% and most preferably about 2.5 mol% and the ratio of hydrogen to ethylene is preferably 100 to 400 mol/kmol, more preferably 150 to 300 mol/kmol and most preferably about 210 mol/kmol.

The comonomer to ethylene ratio has influence on the desired density of the bimodal polymer. Hence, it is preferred that the ratio of comonomer to ethylene is 20 to 150 mol/kmol, more preferably 50 to 100 mol/kmol and most preferably about 80 mol/kmol. Preferably, after the polymer is withdrawn from the gas-

phase reactor and then mixed with further additives as anti-oxidants and/or process stabilizers by blending.

The polymer mix of polymer (A) and polyolefin (B) is then blended with filler (C) and with any suitable method known in the art. These methods include compounding in a twin-screw extruder, like a counter-rotating twin-screw extruder or a co-rotating twin-screw extruder and compounding in a single-screw extruder.

In addition, the present invention comprises a new multi-layer material comprising at least

- a) a substrate as a first layer (I) and
- b) a multimodal polymer composition as described above as at least one further layer (II).

Preferably, the multi-layer material consists of

- a) a substrate as a first layer (I) and
- b) a multimodal polymer composition as described above as at least one further layer (II).

It is further preferred that the multi-layer material is a two-layer or three-layer material consisting of a substrate as a first layer and of a polymer composition for the second and third layer, whereby preferably at least the second layer is a polymer composition as defined above. The layers can of course be in any order. Optionally, this multi-layer material comprises adhesion promoters as tetra-isopropyl titanate, tetra-stearyl titanate, tetrakis(2-ethylhexyl) titanate, poly(dibutyltitanate).

Preferably, the substrate is selected from the group consisting of paper, paperboard, aluminium film and plastic film.

Preferably, the multi-layer material comprises as a further layer (III) a low density polyethylene (LDPE). Thereby, it is preferred that the low density polyethylene has a density of 900 to 950 kg/m³, more preferably from 915 to 925 kg/m³. In addition, it is preferred that the melt flow rate MFR₂ of the low density polyethylene (LDPE) is of 2.0 to 20.0 g/10 min, more preferably from 3.0 to 10.0 g/10 min.

Preferably, the coating weight of layer (II) comprising the polymer composition according to the present invention ranges from 5 to 60 g/m² and more preferably from 10 to 45 g/m². Additionally, it is preferred that the layer (III) comprising a low density polyethylene (LDPE) as described above has a coating weight of 0 to 25, more preferably from 3 to 18 g/m².

The present invention also comprises a film, preferably a cast film, comprising the multimodal polymer composition as described above, more preferably, the film consists of the multimodal polymer composition of the present invention.

Furthermore, the present invention provides a process for producing a multi-layer material comprising the inventive polymer composition as described above. Thereby, it is preferred that the multimodal polymer composition as described above is applied on a substrate by a film-coating line comprising an unwind, a wind, a chill roll and a coating die. Preferably, the speed of the coating line ranges from 50 to 5000 m/min, more preferably from 100 to 1500 m/min. The coating may be done as any coating line known in the art. It is preferred to employ a coating line with at least two extruders to make it possible to produce multilayered coatings with different polymers. It is also possible to have arrange-

ments to treat the polymer melt exiting the die to improve adhesion, e.g. by ozone treatment, corona treatment or flame treatment.

In addition, the present invention comprises the use of the multimodal polymer composition as defined above for extrusion coating, in particular for extrusion coating producing a multi-layer material as described above.

Furthermore, the present invention relates to the use of the multimodal polymer composition for films, preferably cast films.

In the following the present invention is demonstrated by means of examples.

Examples

Measurements

WVTR:

Water vapor transmission rate was measured at 90 % relative humidity and 38 °C temperature according to the method ASTM E96.

Basis weight or coating weight:

Basis weight (or coating weight) was determined as follows: Five samples were cut off from the extrusion coated paper parallel in the transverse direction of the line. The size of the samples was 10 cm × 10 cm. The samples were dried in an oven at 105 °C for one hour. The samples were then weighed and the coating weight was calculated as the difference between the basis weight of the coated structure and the basis weight of the substrate. The result was given as a weight of the plastic per square meter.

Molecular weight averages and molecular weight distribution:

Molecular weight averages and molecular weight distribution were determined by ISO 16014, part 2 universal calibration (narrow MWD polystyrene standards (universal alibration) and a set of 2 x mixed bed + 1 x 10⁷ Å Tosohas (JP) columns were used).

Density:

Density was determined according to ISO 1183-1987.

Melt flow rate or melt index:

Melt flow rate (also referred to as melt index) was determined according to ISO 1133, at 190 °C. The load used in the measurement is indicated as a subscript, i.e. MFR₂ denotes the MFR measured under 2.16 kg load.

Flow rate ratio:

Flow rate ratio is a ratio of two melt flow rates measured for the same polymer under two different loads. The loads are indicated as a subscript, i.e., FRR_{5/2} denotes the ratio of MFR₅ to MFR₂.

Curling:

Curling was determined by cutting a circular sample having an area of 100 cm² within two hours after the coating. The sample is then allowed freely to curl at the table for two minutes. The curl is then measured as the difference (in mm) from the table to the curled sheet.

Example 1

A dry blend of pellets was made of 650 kg of the low density polyethylene CA8200 of 300 kg of a talc filler Finntalc MO5SL, manufactured and sold by Mondo Minerals and 50 kg of Clariant PP6100 PP wax. This dry blend was then compounded and pelletized by using the above-mentioned ZSK70 extruder. The melt temperature during the extrusion was 200°C. The composition was then dried at 60°C for 6 hours to remove the moisture. CA8200 is a low density polyethylene designed for extrusion coating, produced and marketed by Borealis. It is produced by free radical polymerization in a high pressure autoclave process. It has an MFR₂ of 7.5 g/10 min and a density of 920 kg/m³. Clariant PP6100 is a low molecular weight propylene polymer having a number average molecular weight of 2,090 g/mol, weight average molecular weight 5,370 g/mol, z-average molecular weight 10,900 g/mol and melting temperature in DSC analysis 109°C. The composition had a density of 1,195.7 kg/m³ and MFR₂ of 6.1 g/10 min.

Comparative Example 1

The procedure of Example 1 was repeated, except that the amount of CA8200 was 700 kg and Clariant PP6100 was not used. Moreover, no drying at 60°C was done.

Table 1: Data for compositions containing polyolefin and talc used in cast films.

Example	Composition	MFR ₂ g/10 min	Density 920 kg/m ³
Example 1	LD/PP/talc	NA	NA
Comparative Example 1	LD/ - /talc	NA	NA

Example 2

The composition of Example 1 was used to make a cast film on Collin laboratory scale cast film line, having a single screw extruder with a screw diameter of 30 mm and length to diameter (L/D) ratio of 30. The line speed was about 10 m/s (from 8.9 to 10.3 m/s), the output about 5 kg/h (from 4.91 to 6.07 kg/h), the die temperature 250°C and melt temperature 245°C. The temperature of the chill roll was about 70°C (68 to 72°C). The data can be found in Table 2.

The thickness of the film was 45 μm . The WVTR was 5.0 $\text{g/m}^2/24 \text{ h}$.

Example 3

The procedure of Example 2 was repeated, except that the thickness of the film was 98 μm . The WVTR was 2.3 $\text{g/m}^2/24 \text{ h}$.

Comparative Example 2

The procedure of Example 3 was repeated, except that the composition of Comparative Example 1 was used in place of the composition of Example 1. Data can be found in Table 2.

Table 2: Cast film data.

Example	Composition	Thickness μm	WVTR $\text{g/m}^2/24 \text{ h}$
Example 2	LD/ PP/ talc	45	5.0
Example 3	LD/ PP/ talc	98	2.3
Comparative Example 2	LD/ - / talc	102	2.7

Claims

1. A multimodal polymer composition comprising
 - a. at least one polymer (A) having a weight average molecular weight (M_w) of lower than 60000 g/mol;
 - b. at least one polyolefin (B) having a higher weight average molecular weight (M_w) than polymer (A); and
 - c. a filler (C), whereby
the polymer composition without filler (C) has a density of 940 kg/m³ or lower.
2. A polymer composition according to claim 1 characterized in that at least one polymer (A) is
 - (1) a polyolefin having a weight average molecular weight (M_w) of 10000 to 60000 g/mol, or
 - (2) a wax having weight average molecular weight (M_w) of less than 10000 g/mol.
3. A polymer composition according to claim 2 characterized in that the polyolefin (1) is a low density polyethylene (LDPE), a linear low density polyethylene (LLDPE) or a linear medium density polyethylene (LMDPE).
4. A polymer composition according to claim 2 or 3 characterized in that the wax (2) is selected from one or more of
 - (2a) a polypropylene wax having weight average molecular weight (M_w) of less than 10000 g/mol or a polypropylene wax having weight average

molecular weight (M_w) of less than 10000 g/mol, or

(2b) an alkyl ketene dimer wax having weight average molecular weight (M_w) of less than 10000 g/mol.

5. A polymer composition according to any one of the preceding claims 2 to 4 characterized in that the composition comprises a polyolefin (1) as polymer (A) and a wax (2) as a further polymer (A).
6. A polymer composition according to any one of the preceding claims 1 to 5 characterized in that the polymer (A) has a density of lower than 945 kg/m³.
7. A polymer composition according to any one of the preceding claims 1 to 6 characterized in that the multimodal polymer composition is at least a bimodal polymer composition.
8. A polymer composition according to any one of the preceding claims 1 to 7 characterized in that the polyolefin (B) has a weight average molecular weight (M_w) of higher than 80000 g/mol.
9. A polymer composition according to any one of the preceding claims 1 to 8 characterized in that the polyolefin (B) is a polyethylene.
10. A polymer composition according to claim 9 characterized in that the polyolefin (B) is a low density polyethylene (LDPE), a linear low density polyethylene (LLDPE) or linear medium density polyethylene (LMDPE).

11. A polymer composition according to any one of the preceding claims 1 to 10 characterized in that the total polymer composition comprises 1 to 50 wt% of polymer (A), 40 to 90 wt% of polyolefin (B) and 1 to 50 wt% of filler (C).
12. A polymer composition according to any one of the preceding claims 1 to 11 characterized in that the polymer composition without filler (C) has melt flow rate MFR_2 , according to ISO 1133, at 190 °C, of 5 to 20 g/10min.
13. A polymer composition according to any one of the preceding claims 1 to 12 characterized in that the polymer composition without filler (C) has melt flow rate MFR_5 , according to ISO 1133, at 190 °C, of 20 to 40 g/10min.
14. A polymer composition according to any one of the preceding claims 1 to 13 characterized in that the polymer composition without filler (C) has melt flow ratio MFR_5/MFR_2 of 2.5 to 4.5.
15. A polymer composition according to any one of the preceding claims 1 to 14 characterized in that the polymer composition without filler (C) has a ratio of the weight average molecular weight (M_w) to the number average molecular weight (M_n) from 8 to 25.
16. A polymer composition according to any one of the preceding claims 1 to 15 characterized in that 95 wt% of the filler (C) has a particle size of less than 10 μm .

17. A polymer composition according to any one of the preceding claims 1 to 16 characterized in that the filler (C) is talc.
18. A polymer composition according to any one of the preceding claims 1 to 17 characterized in that the polymer composition comprises additionally antioxidants(s) and/or process stabilizers of less than 2000 ppm in the total composition.
19. A polymer composition according to any one of the preceding claims 1 to 18 characterized in that the polymer composition is a linear low density polyethylene (LLDPE) or a liner medium density polyethylene (LMDPE), whereby polymer (A) and polyolefin (B) are produced in a multi step polymerization process.
20. A polymer composition according to claim 19 characterized in that the amount of comonomer units in a linear low density polyethylene (LLDPE) or a in liner medium density polyethylene (LMDPE) is 0.1 to 1.0 mol %.
21. A polymer composition according to claim 19 or 20 characterized in that the polymer (A) and the polyolefin (B) is a linear low density polyethylene (LLDPE) or a liner medium density polyethylene (LMDPE), whereby the comonomer units are selected from the group consisting of C₃ α -olefin, C₄ α -olefin, C₅ α -olefin, C₆ α -olefin, C₇ α -olefin, C₈ α -olefin, C₉ α -olefin, C₁₀ α -olefin, C₁₁ α -olefin, C₁₂ α -olefin, C₁₃ α -olefin, C₁₄ α -olefin, C₁₅ α -olefin, C₁₆ α -olefin, C₁₇ α -olefin, C₁₈ α -olefin, C₁₉ α -olefin and C₂₀ α -olefin.

22. A polymer composition according to any one of the preceding claims 1 to 18 characterized in that the polymer (A) is a wax (2) according to claim (4) and the polyolefin (B) is a linear low density polyethylene (LLDPE) or low density polyethylene (LDPE).
23. A polymer composition according to claim 22 characterized in that the polymer composition comprises additionally a polyolefin (1) being a linear low density polyethylene (LLDPE) as a further polymer (A).
24. A polymer composition according to claim 20 or 21 characterized in that the polymer composition is a linear low density polyethylene (LLDPE) whereby polyolefin (1) (polymer (A)) being a linear low density polyethylene (LLDPE) is the lower molecular weight fraction of LLDPE and polyolefin (B) being a linear low density polyethylene (LLDPE) is the higher molecular weight fraction of the LLDPE.
25. A polymer composition according to claim 24 characterized in that the polymer (A) and polyolefin (B) are a mechanical blend, preferably an in-situ blend produced in a multistage polymerization process.
26. A multi-layer material comprising
 - a. a substrate as a first layer (I)
 - b. a multimodal polymer composition according to any one of the preceding claims as at least a further layer (II).

27. A multi-layer material according to claim 26 characterized in that the substrate is selected from the group consisting of paper, paperboard, aluminum film and plastic film.
28. A multi-layer material according to claim 26 or 27 characterized in that the multi-layer material comprises as a further layer (III) comprising a low density polyethylene (LDPE).
29. A multi-layer material according to any one of the preceding claims 26 to 28 characterized in that the low density polyethylene (LDPE) layer (III) has a melt flow rate MFR_2 , according to ISO 1133, at 190°C, of at least 5 g/10 min.
30. A film comprising a multimodal polymer composition according to any one of the preceding claims 1 to 25.
31. A process for producing a composition according to any one of the preceding claims 1 to 25 characterized in that
 - a. polymer (A) and polyolefin (B) are produced together in a multi-stage process comprising a loop reactor and a gas phase reactor, wherein polymer (A) is generated in at least one loop reactor and the polyolefin (B) is generated in a gas phase reactor; and
 - b. filler (C) and the composition comprising polymer (A) and polyolefin (B) are blended together and compounded.
32. A process for producing a composition according to claim 31 characterized in that the catalyst used for the process producing the composition comprising polymer (A) and polyolefin (B) is a high activity procatalyst comprising a particulate inorganic support, a chlorine compound deposited on the support, wherein the chlorine compound is the same as or different from the titanium compound, whereby the inorganic support is contacted with an alkyl metal chloride which is

soluble in non-polar hydrocarbon solvents, and has the formula $R_n\text{MECL}_3$.
 $n)_m$ wherein R is a C_1 - C_{20} alkyl group, Me is a metal of group III(13) of the periodic table, $n=1$ or 2 and $m=1$ or 2, to give a first reaction product, and

the first reaction product is contacted with a compound containing hydrocarbyl and hydrocarbyl oxide linked to magnesium which is soluble in non-polar hydrocarbon solvents, to give a second reaction product, and the first reaction product is contacted with a compound containing hydrocarbyl and hydrocarbyl oxide linked to magnesium which is soluble in non-polar hydrocarbon solvents, to give a second reaction product, and the second reaction product is contacted with a titanium compound which contains chlorine, having the formula $\text{Cl}_x\text{Ti}(\text{OR}^{\text{IV}})_{4-x}$ wherein R^{IV} is a C_2 - C_{20} hydrocarbyl group and x is 3 or 4, to give the procatalyst.

33. A process for producing a multi-layer material according to any one of the claims 26 to 29 characterized in that the multimodal polymer composition according to any one of the claims 1 to 25 is applied on the substrate by a film coating line comprising an unwind, a wind, a chill roll and a coating die.
34. Use of the multimodal polymer composition according to any one of the claims 1 to 25 for extrusion coating.
35. Use according to claim 34 characterized in that the polymer extrusion composition according to any one of the claims 1 to 25 is used for extrusion coating producing a multi-layer material according to any one of the claim 26 to 29.
36. Use of the multimodal polymer composition according to any one of the claims 1 to 25 for a film, preferably for a cast film.

PATENT COOPERATION TREATY

From the RECEIVING OFFICE

PCT

To:

KADOR & PARTNER
Corneliusstrasse 15
D-80469 München
ALLEMAGNE

EINGEGANGEN

24 Jan. 2005

KADOR & PARTNER

NOTIFICATION OF THE INTERNATIONAL
APPLICATION NUMBER AND OF THE
INTERNATIONAL FILING DATE

(PCT Rule 20.5(c))

Date of mailing
(day/month/year)

21 JAN 2005

Applicant's or agent's file reference

K 50 783/7nd

IMPORTANT NOTIFICATION

International application No.

PCT/EP2005/000221

International filing date (day/month/year)

12/01/2005

Priority date (day/month/year)

Applicant

BOREALIS TECHNOLOGY OY

Title of the invention

1. The applicant is hereby notified that the international application has been accorded the international application number and the international filing date indicated above.
2. The applicant is further notified that the record copy of the international application was transmitted to the International Bureau on the above date of mailing.

3.

☐

Other: _____

* The International Bureau monitors the transmittal of the record copy by the receiving Office and will notify the applicant (with Form PCT/IB/301) of its receipt. Should the record copy not have been received by the expiration of 14 months from the priority date, the International Bureau will notify the applicant (Rule 22.1(c)).

Name and mailing address of the Receiving Office



European Patent Office, P.B. 5818 Patentlaan 2
NL-2280 HV Rijswijk
Tel. (+31-70) 340-2040
Fax: (+31-70) 340-3016

Authorized officer

PCT

REQUEST

The undersigned requests that the present international application be processed according to the Patent Cooperation Treaty.

For receiving Office use only

PCT/EP 2005 / 0 0 0 2 2 1

International Application No.

(12 01 2005)

International Filing Date

12 JAN 2005

EUROPEAN PATENT OFFICE

PCT INTERNATIONAL APPLICATION

Name of receiving Office and "PCT International Application"

Applicant's or agent's file reference
(if desired) (12 characters maximum) K 50 783/7nd

Box No. I TITLE OF INVENTION

Extrusion coating polyethylene

Box No. II APPLICANT

☐ This person is also inventor

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

Borealis Technology Oy
P.O. Box 330
FIN-06101 Porvoo
Finland

Telephone No.

Facsimile No.

Teleprinter No.

Applicant's registration No. with the Office

State (that is, country) of nationality:

FI

State (that is, country) of residence:

FI

This person is applicant
for the purposes of:

☐ all designated
States

☒ all designated States except
the United States of America

☐ the United States
of America only

☐ the States indicated in
the Supplemental Box

Box No. III FURTHER APPLICANT(S) AND/OR (FURTHER) INVENTOR(S)

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

LAIHO, Erkki
Kaivantotie 7
06450 Porvoo
Finland

This person is:

☐ applicant only

☒ applicant and inventor

☐ inventor only (If this check-box is
marked, do not fill in below.)

Applicant's registration No. with the Office

State (that is, country) of nationality:

FI

State (that is, country) of residence:

FI

This person is applicant
for the purposes of:

☐ all designated
States

☐ all designated States except
the United States of America

☒ the United States
of America only

☐ the States indicated in
the Supplemental Box

☒ Further applicants and/or (further) inventors are indicated on a continuation sheet.

Box No. IV AGENT OR COMMON REPRESENTATIVE; OR ADDRESS FOR CORRESPONDENCE

The person identified below is hereby/has been appointed to act on behalf of the applicant(s) before the competent International Authorities as:

☒ agent

☐ common
representative

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)

Kador & Partner
Corneliusstr. 15
80469 Munich
Germany

Telephone No.

+49-89-2015252

Facsimile No.

+49-89-2015242

Teleprinter No.

Agent's registration No. with the Office

☐ Address for correspondence: Mark this check-box where no agent or common representative is/has been appointed and the space above is used instead to indicate a special address to which correspondence should be sent.

Continuation of Box No. III FURTHER APPLICANT(S) AND/OR (FURTHER) INVENTOR(S)

If none of the following sub-boxes is used, this sheet should not be included in the request.

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

SAINIO, Markku
Myrskyläntie 696
06100 Porvoo
Finland

This person is:

- ☐ applicant only
☒ applicant and inventor
☐ inventor only (If this check-box is marked, do not fill in below.)

Applicant's registration No. with the Office

State (that is, country) of nationality:

FI

State (that is, country) of residence:

FI

This person is applicant for the purposes of:

☐ all designated States☐ all designated States except the United States of America☒ the United States of America only☐ the States indicated in the Supplemental Box

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

VÄHÄLÄ, Martti
Vikkulankatu 49 A 1
37150 Nokia
Finland

This person is:

- ☐ applicant only
☒ applicant and inventor
☐ inventor only (If this check-box is marked, do not fill in below.)

Applicant's registration No. with the Office

State (that is, country) of nationality:

FI

State (that is, country) of residence:

FI

This person is applicant for the purposes of:

☐ all designated States☐ all designated States except the United States of America☒ the United States of America only☐ the States indicated in the Supplemental Box

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

This person is:

- ☐ applicant only
☐ applicant and inventor
☐ inventor only (If this check-box is marked, do not fill in below.)

Applicant's registration No. with the Office

State (that is, country) of nationality:

State (that is, country) of residence:

This person is applicant for the purposes of:

☐ all designated States☐ all designated States except the United States of America☐ the United States of America only☐ the States indicated in the Supplemental Box

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

This person is:

- ☐ applicant only
☐ applicant and inventor
☐ inventor only (If this check-box is marked, do not fill in below.)

Applicant's registration No. with the Office

State (that is, country) of nationality:

State (that is, country) of residence:

This person is applicant for the purposes of:

☐ all designated States☐ all designated States except the United States of America☐ the United States of America only☐ the States indicated in the Supplemental Box☐ Further applicants and/or (further) inventors are indicated on another continuation sheet.

Box No. V DESIGNATIONS

The filing of this request constitutes under Rule 4.9(a), the designation of all Contracting States bound by the PCT on the international filing date, for the grant of every kind of protection available and, where applicable, for the grant of both regional and national patents.

However,

- ☐ DE Germany is not designated for any kind of national protection
- ☐ KR Republic of Korea is not designated for any kind of national protection
- ☐ RU Russian Federation is not designated for any kind of national protection

(The check-boxes above may be used to exclude (irrevocably) the designations concerned in order to avoid the ceasing of the effect, under the national law, of an earlier national application from which priority is claimed. See the Notes to Box No. V as to the consequences of such national law provisions in these and certain other States.)

Box No. VI PRIORITY CLAIM

The priority of the following earlier application(s) is hereby claimed:

Filing date of earlier application (day/month/year)	Number of earlier application	Where earlier application is:		
		national application: country or Member of WTO	regional application:* regional Office	international application: receiving Office
item (1)				
item (2)				
item (3)				

☐ Further priority claims are indicated in the Supplemental Box.

The receiving Office is requested to prepare and transmit to the International Bureau a certified copy of the earlier application(s) (only if the earlier application was filed with the Office which for the purposes of this international application is the receiving Office) identified above as:

☐ all items ☐ item (1) ☐ item (2) ☐ item (3) ☐ other, see Supplemental Box

* Where the earlier application is an ARIPO application, indicate at least one country party to the Paris Convention for the Protection of Industrial Property or one Member of the World Trade Organization for which that earlier application was filed (Rule 4.10(b)(ii)):

Box No. VII INTERNATIONAL SEARCHING AUTHORITY

Choice of International Searching Authority (ISA) (if two or more International Searching Authorities are competent to carry out the international search, indicate the Authority chosen; the two-letter code may be used):

ISA / .EP.....

Request to use results of earlier search; reference to that search (if an earlier search has been carried out by or requested from the International Searching Authority):

Date (day/month/year) Number Country (or regional Office)

Box No. VIII DECLARATIONS

The following declarations are contained in Boxes Nos. VIII (i) to (v) (mark the applicable check-boxes below and indicate in the right column the number of each type of declaration):

Number of
declarations

- ☐ Box No. VIII (i) Declaration as to the identity of the inventor :
- ☐ Box No. VIII (ii) Declaration as to the applicant's entitlement, as at the international filing date, to apply for and be granted a patent :
- ☐ Box No. VIII (iii) Declaration as to the applicant's entitlement, as at the international filing date, to claim the priority of the earlier application :
- ☐ Box No. VIII (iv) Declaration of inventorship (only for the purposes of the designation of the United States of America) :
- ☐ Box No. VIII (v) Declaration as to non-prejudicial disclosures or exceptions to lack of novelty :

Box No. IX CHECK LIST; LANGUAGE OF FILING

This international application contains:		This international application is accompanied by the following item(s) (mark the applicable check-boxes below and indicate in right column the number of each item):		Number of items
(a) in paper form, the following number of sheets:		1. <input checked="" type="checkbox"/> fee calculation sheet		1
request (including declaration sheets)	4	2. <input checked="" type="checkbox"/> original separate power of attorney		follows
description (excluding sequence listing and/or tables related thereto)	33	3. <input type="checkbox"/> original general power of attorney		
claims	7	4. <input checked="" type="checkbox"/> copy of general power of attorney; reference number, if any: 42788.		1
abstract	1	5. <input type="checkbox"/> statement explaining lack of signature		
drawings		6. <input type="checkbox"/> priority document(s) identified in Box No. VI as item(s):		
Sub-total number of sheets	45	7. <input type="checkbox"/> translation of international application into (language):		
sequence listing		8. <input type="checkbox"/> separate indications concerning deposited microorganism or other biological material		
tables related thereto		9. <input type="checkbox"/> sequence listing in computer readable form (indicate type and number of carriers)		
(for both, actual number of sheets if filed in paper form, whether or not also filed in computer readable form; see (c) below)		(i) <input type="checkbox"/> copy submitted for the purposes of international search under Rule 13ter only (and not as part of the international application):		
Total number of sheets	45	(ii) <input type="checkbox"/> (only where check-box (b)(i) or (c)(i) is marked in left column) additional copies including, where applicable, the copy for the purposes of international search under Rule 13ter		
(b) <input type="checkbox"/> only in computer readable form (Section 801(a)(i))		(iii) <input type="checkbox"/> together with relevant statement as to the identity of the copy or copies with the sequence listing mentioned in left column		
(i) <input type="checkbox"/> sequence listing		10. <input type="checkbox"/> tables in computer readable form related to sequence listing (indicate type and number of carriers)		
(ii) <input type="checkbox"/> tables related thereto		(i) <input type="checkbox"/> copy submitted for the purposes of international search under Section 802(b-quater) only (and not as part of the international application)		
(c) <input type="checkbox"/> also in computer readable form (Section 801(a)(ii))		(ii) <input type="checkbox"/> (only where check-box (b)(ii) or (c)(ii) is marked in left column) additional copies including, where applicable, the copy for the purposes of international search under Section 802(b-quater)		
(i) <input type="checkbox"/> sequence listing		(iii) <input type="checkbox"/> together with relevant statement as to the identity of the copy or copies with the tables mentioned in left column		
(ii) <input type="checkbox"/> tables related thereto		11. <input type="checkbox"/> other (specify):		1
Type and number of carriers (diskette, CD-ROM, CD-R or other) on which are contained the				
<input type="checkbox"/> sequence listing:				
<input type="checkbox"/> tables related thereto:				
(additional copies to be indicated under items 9(ii) and/or 10(ii), in right column)				

Figure of the drawings which should accompany the abstract:

Language of filing of the international application:

EN

Box No. X SIGNATURE OF APPLICANT, AGENT OR COMMON REPRESENTATIVE

Next to each signature, indicate the name of the person signing and the capacity in which the person signs (if such capacity is not obvious from reading the request).



Dr. U. Kador

For receiving Office use only			
1. Date of actual receipt of the purported international application:	12.01.05	12 JAN 2005	2. Drawings: <input type="checkbox"/> received: <input type="checkbox"/> not received:
3. Corrected date of actual receipt due to later but timely received papers or drawings completing the purported international application:			
4. Date of timely receipt of the required corrections under PCT Article 11(2):			
5. International Searching Authority (if two or more are competent): ISA /	6. <input type="checkbox"/> Transmittal of search copy delayed until search fee is paid		

For International Bureau use only

Date of receipt of the record copy by the International Bureau:

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference K 50 783/7nd	FOR FURTHER ACTION see Form PCT/ISA/220 as well as, where applicable, item 5 below.	
International application No. PCT/EP2005/000221	International filing date (day/month/year) 12/01/2005	(Earliest) Priority Date (day/month/year)
Applicant BOREALIS TECHNOLOGY OY		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 5 sheets.



It is also accompanied by a copy of each prior art document cited in this report.

1. **Basis of the report**

- a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.



The international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).



With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, see Box No. I.



Certain claims were found unsearchable (See Box II).



Unity of invention is lacking (see Box III).

4. With regard to the **title**,



the text is approved as submitted by the applicant.



the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,



the text is approved as submitted by the applicant.



the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box No. IV. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. With regard to the **drawings**,

- a. the figure of the **drawings** to be published with the abstract is Figure No. _____



as suggested by the applicant.



as selected by this Authority, because the applicant failed to suggest a figure.



as selected by this Authority, because this figure better characterizes the invention.



none of the figures is to be published with the abstract.

Abstract

The present invention relates to a polymer composition with good chemical properties and barrier properties being multimodal and comprising a polymer (A) having a weight average molecular weight of lower than 60000 g/mol and a polyolefin (B) having a higher weight average molecular weight than polymer (A) and filler (C), whereby a polymer composition without filler (C) has a density of 940kg/m³ or lower.

INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP2005/000221

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C08F297/08 C08L23/08 C08L23/12

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08F C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 1 375 584 A (BOREALIS TECHNOLOGY OY) 2 January 2004 (2004-01-02) claim 1	1-36
A	EP 1 146 078 A (BOREALIS TECHNOLOGY OY) 17 October 2001 (2001-10-17) paragraphs '0010!', '0015!', '0016!', '0025!', '0027! claims 1,5,9,11,15	1-36
A	US 2004/034169 A1 (ZHOU ZHIQIANG JIMMY ET AL) 19 February 2004 (2004-02-19) paragraphs '0034!', '0038!', '0051! claims 1,12	1-36
	----- -/--	

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search

20 July 2005

Date of mailing of the international search report

29/07/2005

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
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Fax: (+31-70) 340-3016

Authorized officer

Lippert, S

INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP2005/000221

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 2003/171501 A1 (KALLIO KALLE ET AL) 11 September 2003 (2003-09-11) paragraphs '0029!, '0032!, '0036!, '0051!, '0053!, '0054! example 3; table 1 claim 1	1-36
A	----- US 2003/149180 A1 (VAN DUN JOZEF J ET AL) 7 August 2003 (2003-08-07) claim 1 -----	1-36

INTERNATIONAL SEARCH REPORT

information on patent family members

International Application No

PCT/EP2005/000221

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
EP 1375584	A	02-01-2004	EP 1375584 A1	02-01-2004
			AU 2003239634 A1	06-01-2004
			BR 0311562 A	26-04-2005
			WO 2004000933 A1	31-12-2003
EP 1146078	A	17-10-2001	EP 1146078 A1	17-10-2001
			AU 5472101 A	30-10-2001
			BR 0110067 A	31-12-2002
			CN 1422302 A	04-06-2003
			WO 0179347 A2	25-10-2001
			PL 358104 A1	09-08-2004
			US 2003149162 A1	07-08-2003
US 2004034169	A1	19-02-2004	AU 2003240541 A1	19-12-2003
			BR 0311703 A	08-03-2005
			CA 2487931 A1	11-12-2003
			EP 1511805 A1	09-03-2005
			WO 03102075 A1	11-12-2003
US 2003171501	A1	11-09-2003	AU 6622001 A	14-01-2002
			CN 1440330 A	03-09-2003
			EP 1294564 A1	26-03-2003
			WO 0202323 A1	10-01-2002
			ZA 200300246 A	21-01-2004
US 2003149180	A1	07-08-2003	US 2004198911 A1	07-10-2004
			BR 0211959 A	21-09-2004
			CA 2459552 A1	27-02-2003
			CN 1543483 A	03-11-2004
			EP 1417260 A1	12-05-2004
			JP 2005500423 T	06-01-2005
			MX PA04001496 A	14-05-2004
			WO 03016396 A1	27-02-2003

INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP2005/000221

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C08F297/08 C08L23/08 C08L23/12

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08F C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 1 375 584 A (BOREALIS TECHNOLOGY OY) 2 January 2004 (2004-01-02) claim 1	1-36
A	EP 1 146 078 A (BOREALIS TECHNOLOGY OY) 17 October 2001 (2001-10-17) paragraphs '0010!', '0015!', '0016!', '0025!', '0027! claims 1,5,9,11,15	1-36
A	US 2004/034169 A1 (ZHOU ZHIQIANG JIMMY ET AL) 19 February 2004 (2004-02-19) paragraphs '0034!', '0038!', '0051! claims 1,12	1-36
	----- -/--	

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

A document defining the general state of the art which is not considered to be of particular relevance

E earlier document but published on or after the international filing date

L document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

O document referring to an oral disclosure, use, exhibition or other means

P document published prior to the international filing date but later than the priority date claimed

T later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

X document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

Y document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

& document member of the same patent family

Date of the actual completion of the international search

20 July 2005

Date of mailing of the international search report

29/07/2005

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax: (+31-70) 340-3016

Authorized officer

Lippert, S

INTERNATIONAL SEARCH REPORT

International Application No.
PCT/EP2005/000221

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 2003/171501 A1 (KALLIO KALLE ET AL) 11 September 2003 (2003-09-11) paragraphs '0029!, '0032!, '0036!, '0051!, '0053!, '0054! example 3; table 1 claim 1	1-36
A	US 2003/149180 A1 (VAN DUN JOZEF J ET AL) 7 August 2003 (2003-08-07) claim 1	1-36

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP2005/000221

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
EP 1375584	A	02-01-2004	EP 1375584 A1	02-01-2004
			AU 2003239634 A1	06-01-2004
			BR 0311562 A	26-04-2005
			WO 2004000933 A1	31-12-2003
EP 1146078	A	17-10-2001	EP 1146078 A1	17-10-2001
			AU 5472101 A	30-10-2001
			BR 0110067 A	31-12-2002
			CN 1422302 A	04-06-2003
			WO 0179347 A2	25-10-2001
			PL 358104 A1	09-08-2004
			US 2003149162 A1	07-08-2003
US 2004034169	A1	19-02-2004	AU 2003240541 A1	19-12-2003
			BR 0311703 A	08-03-2005
			CA 2487931 A1	11-12-2003
			EP 1511805 A1	09-03-2005
			WO 03102075 A1	11-12-2003
US 2003171501	A1	11-09-2003	AU 6622001 A	14-01-2002
			CN 1440330 A	03-09-2003
			EP 1294564 A1	26-03-2003
			WO 0202323 A1	10-01-2002
			ZA 200300246 A	21-01-2004
US 2003149180	A1	07-08-2003	US 2004198911 A1	07-10-2004
			BR 0211959 A	21-09-2004
			CA 2459552 A1	27-02-2003
			CN 1543483 A	03-11-2004
			EP 1417260 A1	12-05-2004
			JP 2005500423 T	06-01-2005
			MX PA04001496 A	14-05-2004
			WO 03016396 A1	27-02-2003

PATENT COOPERATION TREATY

From the INTERNATIONAL SEARCHING AUTHORITY

PCT

To:

KADOR & PARTNER
Corneliusstrasse 15
D-80469 München
GERMANY

NOTIFICATION OF TRANSMITTAL OF
THE INTERNATIONAL SEARCH REPORT AND
THE WRITTEN OPINION OF THE INTERNATIONAL
SEARCHING AUTHORITY, OR THE DECLARATION

(PCT Rule 44.1)

Date of mailing (day/month/year) 29/07/2005	
Applicant's or agent's file reference K 50 783/7nd	FOR FURTHER ACTION See paragraphs 1 and 4 below
International application No. PCT/EP2005/000221	International filing date (day/month/year) 12/01/2005
Applicant BOREALIS TECHNOLOGY OY	

1. ☒ The applicant is hereby notified that the international search report and the written opinion of the International Searching Authority have been established and are transmitted herewith.

Filing of amendments and statement under Article 19:

The applicant is entitled, if he so wishes, to amend the claims of the International Application (see Rule 46):

When? The time limit for filing such amendments is normally 2 months from the date of transmittal of the International Search Report; however, for more details, see the notes on the accompanying sheet.

Where? Directly to the International Bureau of WIPO, 34 chemin des Colombettes
1211 Geneva 20, Switzerland, Facsimile No.: (41-22) 740.14.35

For more detailed instructions, see the notes on the accompanying sheet.

2. ☐ The applicant is hereby notified that no international search report will be established and that the declaration under Article 17(2)(a) to that effect and the written opinion of the International Searching Authority are transmitted herewith.

3. ☐ **With regard to the protest** against payment of (an) additional fee(s) under Rule 40.2, the applicant is notified that:

- ☐ the protest together with the decision thereon has been transmitted to the International Bureau together with the applicant's request to forward the texts of both the protest and the decision thereon to the designated Offices.
☐ no decision has been made yet on the protest; the applicant will be notified as soon as a decision is made.

4. **Minors**

Shortly after the expiration of **18 months** from the priority date, the international application will be published by the International Bureau. If the applicant wishes to avoid or postpone publication, a notice of withdrawal of the international application, or of the priority claim, must reach the International Bureau as provided in Rules 90bis.1 and 90bis.3, respectively, before the completion of the technical preparations for international publication.

The applicant may submit comments on an informal basis on the written opinion of the International Searching Authority to the International Bureau. The International Bureau will send a copy of such comments to all designated Offices unless an international preliminary examination report has been or is to be established. These comments would also be made available to the public but not before the expiration of 30 months from the priority date.

Within **19 months** from the priority date, but only in respect of some designated Offices, a demand for international preliminary examination must be filed if the applicant wishes to postpone the entry into the national phase until **30 months** from the priority date (in some Offices even later); otherwise, the applicant must, **within 20 months** from the priority date, perform the prescribed acts for entry into the national phase before those designated Offices.

In respect of other designated Offices, the time limit of **30 months** (or later) will apply even if no demand is filed within 19 months.

See the Annex to Form PCT/IB/301 and, for details about the applicable time limits, Office by Office, see the *PCT Applicant's Guide*, Volume II, National Chapters and the WIPO Internet site.

Name and mailing address of the International Searching Authority



European Patent Office, P.B. 5818 Patentlaan 2
NL-2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax: (+31-70) 340-3016

Authorized officer

Maja Kjellström

NOTES TO FORM PCT/ISA/220

These Notes are intended to give the basic instructions concerning the filing of amendments under article 19. The Notes are based on the requirements of the Patent Cooperation Treaty, the Regulations and the Administrative Instructions under that Treaty. In case of discrepancy between these Notes and those requirements, the latter are applicable. For more detailed information, see also the PCT Applicant's Guide, a publication of WIPO.

In these Notes, "Article", "Rule", and "Section" refer to the provisions of the PCT, the PCT Regulations and the PCT Administrative Instructions respectively.

INSTRUCTIONS CONCERNING AMENDMENTS UNDER ARTICLE 19

The applicant has, after having received the international search report, one opportunity to amend the claims of the international application. It should however be emphasized that, since all parts of the international application (claims, description and drawings) may be amended during the international preliminary examination procedure, there is usually no need to file amendments of the claims under Article 19 except where, e.g. the applicant wants the latter to be published for the purposes of provisional protection or has another reason for amending the claims before international publication. Furthermore, it should be emphasized that provisional protection is available in some States only.

What parts of the international application may be amended?

Under Article 19, only the claims may be amended.

During the international phase, the claims may also be amended (or further amended) under Article 34 before the International Preliminary Examining Authority. The description and drawings may only be amended under Article 34 before the International Examining Authority.

Upon entry into the national phase, all parts of the international application may be amended under Article 28 or, where applicable, Article 41.

When?

Within 2 months from the date of transmittal of the international search report or 16 months from the priority date, whichever time limit expires later. It should be noted, however, that the amendments will be considered as having been received on time if they are received by the International Bureau after the expiration of the applicable time limit but before the completion of the technical preparations for international publication (Rule 46.1).

Where not to file the amendments?

The amendments may only be filed with the International Bureau and not with the receiving Office or the International Searching Authority (Rule 46.2).

Where a demand for international preliminary examination has been/is filed, see below.

How?

Either by cancelling one or more entire claims, by adding one or more new claims or by amending the text of one or more of the claims as filed.

A replacement sheet must be submitted for each sheet of the claims which, on account of an amendment or amendments, differs from the sheet originally filed.

All the claims appearing on a replacement sheet must be numbered in Arabic numerals. Where a claim is cancelled, no renumbering of the other claims is required. In all cases where claims are renumbered, they must be renumbered consecutively (Administrative Instructions, Section 205(b)).

The amendments must be made in the language in which the international application is to be published.

What documents must/may accompany the amendments?

Letter (Section 205(b)):

The amendments must be submitted with a letter.

The letter will not be published with the international application and the amended claims. It should not be confused with the "Statement under Article 19(1)" (see below, under "Statement under Article 19(1)").

The letter must be in English or French, at the choice of the applicant. However, if the language of the international application is English, the letter must be in English; if the language of the international application is French, the letter must be in French.

NOTES TO FORM PCT/ISA/220 (continued)

The letter must indicate the differences between the claims as filed and the claims as amended. It must, in particular, indicate, in connection with each claim appearing in the international application (it being understood that identical indications concerning several claims may be grouped), whether

- (i) the claim is unchanged;
- (ii) the claim is cancelled;
- (iii) the claim is new;
- (iv) the claim replaces one or more claims as filed;
- (v) the claim is the result of the division of a claim as filed.

The following examples illustrate the manner in which amendments must be explained in the accompanying letter:

1. [Where originally there were 48 claims and after amendment of some claims there are 51]:
"Claims 1 to 29, 31, 32, 34, 35, 37 to 48 replaced by amended claims bearing the same numbers; claims 30, 33 and 36 unchanged; new claims 49 to 51 added."
2. [Where originally there were 15 claims and after amendment of all claims there are 11]:
"Claims 1 to 15 replaced by amended claims 1 to 11."
3. [Where originally there were 14 claims and the amendments consist in cancelling some claims and in adding new claims]:
"Claims 1 to 6 and 14 unchanged; claims 7 to 13 cancelled; new claims 15, 16 and 17 added." or
"Claims 7 to 13 cancelled; new claims 15, 16 and 17 added; all other claims unchanged."
4. [Where various kinds of amendments are made]:
"Claims 1-10 unchanged; claims 11 to 13, 18 and 19 cancelled; claims 14, 15 and 16 replaced by amended claim 14; claim 17 subdivided into amended claims 15, 16 and 17; new claims 20 and 21 added."

"Statement under article 19(1)" (Rule 46.4)

The amendments may be accompanied by a statement explaining the amendments and indicating any impact that such amendments might have on the description and the drawings (which cannot be amended under Article 19(1)).

The statement will be published with the international application and the amended claims.

It must be in the language in which the international application is to be published.

It must be brief, not exceeding 500 words if in English or if translated into English.

It should not be confused with and does not replace the letter indicating the differences between the claims as filed and as amended. It must be filed on a separate sheet and must be identified as such by a heading, preferably by using the words "Statement under Article 19(1)."

It may not contain any disparaging comments on the international search report or the relevance of citations contained in that report. Reference to citations, relevant to a given claim, contained in the international search report may be made only in connection with an amendment of that claim.

Consequence if a demand for international preliminary examination has already been filed

If, at the time of filing any amendments under Article 19, a demand for international preliminary examination has already been submitted, the applicant must preferably, at the same time of filing the amendments with the International Bureau, also file a copy of such amendments with the International Preliminary Examining Authority (see Rule 62.2(a), first sentence).

Consequence with regard to translation of the international application for entry into the national phase

The applicant's attention is drawn to the fact that, where upon entry into the national phase, a translation of the claims as amended under Article 19 may have to be furnished to the designated/elected Offices, instead of, or in addition to, the translation of the claims as filed.

For further details on the requirements of each designated/elected Office, see Volume II of the PCT Applicant's Guide.

PATENT COOPERATION TREATY

From the
INTERNATIONAL SEARCHING AUTHORITY

To:

see form PCT/ISA/220

PCT

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY (PCT Rule 43bis.1)

Date of mailing
(day/month/year) see form PCT/ISA/210 (second sheet)

Applicant's or agent's file reference
see form PCT/ISA/220

FOR FURTHER ACTION
See paragraph 2 below

International application No.
PCT/EP2005/000221

International filing date (day/month/year)
12.01.2005

Priority date (day/month/year)

International Patent Classification (IPC) or both national classification and IPC
C08F297/08, C08L23/08, C08L23/12

Applicant
BOREALIS TECHNOLOGY OY

1. This opinion contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☒ Box No. II Priority
- ☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☐ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☐ Box No. VII Certain defects in the international application
- ☐ Box No. VIII Certain observations on the international application

2. FURTHER ACTION

If a demand for international preliminary examination is made, this opinion will usually be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA"). However, this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notified the International Bureau under Rule 66.1bis(b) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of three months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.

For further options, see Form PCT/ISA/220.

3. For further details, see notes to Form PCT/ISA/220.

Name and mailing address of the ISA:



European Patent Office
D-80298 Munich
Tel. +49 89 2399 - 0 Tx: 523656 epmu d
Fax: +49 89 2399 - 4465

Authorized Officer

Lippert, S

Telephone No. +49 89 2399-8514



**WRITTEN OPINION OF THE
INTERNATIONAL SEARCHING AUTHORITY**

International application No.
PCT/EP2005/000221

Box No. I Basis of the opinion

1. With regard to the **language**, this opinion has been established on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.
☐ This opinion has been established on the basis of a translation from the original language into the following language , which is the language of a translation furnished for the purposes of international search (under Rules 12.3 and 23.1(b)).
2. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application and necessary to the claimed invention, this opinion has been established on the basis of:
 - a. type of material:
☐ a sequence listing
☐ table(s) related to the sequence listing
 - b. format of material:
☐ in written format
☐ in computer readable form
 - c. time of filing/furnishing:
☐ contained in the international application as filed.
☐ filed together with the international application in computer readable form.
☐ furnished subsequently to this Authority for the purposes of search.
3. ☐ In addition, in the case that more than one version or copy of a sequence listing and/or table relating thereto has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.
4. Additional comments:

Box No. II Priority

1. ☒ The validity of the priority claim has not been considered because the International Searching Authority does not have in its possession a copy of the earlier application whose priority has been claimed or, where required, a translation of that earlier application. This opinion has nevertheless been established on the assumption that the relevant date (Rules 43bis.1 and 64.1) is the claimed priority date.
2. ☐ This opinion has been established as if no priority had been claimed due to the fact that the priority claim has been found invalid (Rules 43bis.1 and 64.1). Thus for the purposes of this opinion, the international filing date indicated above is considered to be the relevant date.
3. Additional observations, if necessary:

**WRITTEN OPINION OF THE
INTERNATIONAL SEARCHING AUTHORITY**

International application No.
PCT/EP2005/000221

Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims	1-36
	No: Claims	
Inventive step (IS)	Yes: Claims	1-36
	No: Claims	
Industrial applicability (IA)	Yes: Claims	1-36
	No: Claims	

2. Citations and explanations

see separate sheet

71/795028

IAP8 Rec'd PCT/PTO 11 JUL 2007

International application No.

**WRITTEN OPINION OF THE
INTERNATIONAL SEARCHING
AUTHORITY (SEPARATE SHEET)**

PCT/EP2005/000221

ad item V:

1. The claimed subject-matter is considered to be novel as none of the prior art documents cited in the International Search Report discloses all the technical features of the present application (Art.33(2) PCT).
2. The claimed subject-matter is considered to be inventive as the specific combination of features as claimed cannot be derived from the prior art documents cited in the International Search Report alone or in combination in an obvious way, nor would any such combination have resulted in the present invention (Art.33(3) PCT).
3. Industrial applicability is given (Art.33(4) PCT).

PATENT COOPERATION TREATY

PCT

NOTIFICATION OF THE RECORDING
OF A CHANGE(PCT Rule 92bis.1 and
Administrative Instructions, Section 422)

From the INTERNATIONAL BUREAU

To:

KADOR & PARTNER
Corneliusstrasse 15
80469 München
Germany

Date of mailing (day/month/year) 08 March 2005 (08.03.2005)	IMPORTANT NOTIFICATION
Applicant's or agent's file reference K 50 783/7nd ✓	
International application No. PCT/EP2005/000221 ✓	International filing date (day/month/year) 12 January 2005 (12.01.2005)

1. The following indications appeared on record concerning:

☒ the applicant ✓ ☒ the inventor ✓ ☐ the agent ☐ the common representative

Name and Address VÄHÄLÄ, Martti Vikkulankatu 49 A 1 FIN-37150 Nokia Finland ✓	State of Nationality FI	State of Residence FI
	Telephone No.	
	Facsimile No.	
	Teleprinter No.	

2. The International Bureau hereby notifies the applicant that the following change has been recorded concerning: ✓

☒ the person ✓ ☐ the name ☐ the address ☐ the nationality ☐ the residence

Name and Address HAAPANIEMI, Karoliina ✓ Dr. Karl-Stainer-Strasse 1-5 ✓ A-6112 Wattens ✓ Austria ✓	State of Nationality FI ✓	State of Residence AT ✓
	Telephone No.	
	Facsimile No.	
	Teleprinter No.	

3. Further observations, if necessary: ✓
Please note that the person in Box 1 has been deleted from the record. And the person in Box 2 has been added to the record as applicant/inventor for US only and inventor for all designated States. ✓

4. A copy of this notification has been sent to:

☒ the receiving Office
☒ the International Searching Authority
☐ the International Preliminary Examining Authority

☐ the designated Offices concerned
☐ the elected Offices concerned
☐ other:

The International Bureau of WIPO
34, chemin des Colombettes
1211 Geneva 20, Switzerland

Authorized officer

Rosana REYES (Fax : 338 89 75)

Attorney Docket No. 37488.01300US

IAP8 Rec'd PCT/PTO 11 JUL 2007

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Application of:

Laiho *et al.*

Application No: Not Yet Assigned

Filed: January 12, 2005

For: EXTRUSION COATING
POLYETHYLENE

)
)
)
)
)
)
)
)
)
)

Group Art Unit: Not Yet Assigned

Examiner: Not Yet Assigned

Mail Stop Amendment

Commissioner of Patents
U.S. Patent and Trademark Office
P.O. Box 1450
Alexandria, VA 22313-1450

In accordance with 37 C.F.R. §§ 1.97 and 1.98, and in compliance with the duty of disclosure set forth in C.F.R. § 1.56, applicants are submitting herewith copy of reference listed on the attached Form PTO/SB/08B for consideration and to be made of record herein by the U.S. Patent and Trademark Office in the above-captioned application. Applicants have also attached a copy of the International Search Report for PCT/EP2005/000221 dated July 20, 2005.

3

Consideration of the foregoing plus the prompt return of a copy of the enclosed Form PTO/SB/08B with the Examiner's initials in the left column in accordance with MPEP 609 are respectfully requested.

Applicants' submission of this Information Disclosure Statement (IDS) shall not be construed as an admission that the cited references are prior art to the present invention, or that the cited references qualify as printed publications. The submission of this IDS also shall not be construed as a representation that a prior art search has been performed.

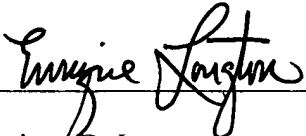
In accordance with 37 C.F.R. § 1.97(b), this Information Disclosure Statement is believed to be submitted before the mailing of a first Official Action. Therefore, it is respectfully submitted that no fee is required for consideration of this information.

However, in the event any fee is deemed necessary, the Commissioner is authorized to charge the undersigned's Deposit Account No. 13-3250 under referenced No. 37488.01300US.

EXCEPT for issue fees payable under 37 C.F.R. § 1.18, the Director is hereby authorized by this paper to charge any additional fees during the entire pendency of this application, including fees due under 37 C.F.R. §§ 1.16 and 1.17 which may be required, including any required extension of time fees, or credit any overpayment to Deposit Account No. 13-3250, reference No. 37488.01300US. This paragraph is intended to be a **CONSTRUCTIVE PETITION FOR EXTENSION OF TIME** in accordance with 37 C.F. R. § 1.136(a)(3).

Respectfully submitted,

**MILBANK, TWEED, HADLEY &
McCLOY LLP**

By: 
Enrique D. Longton
Reg. No. 47,304

Dated: July 11, 2007

Customer No. 38647
MILBANK, TWEED, HADLEY &
McCLOY LLP
1850 K Street, NW
Suite 1100
Washington, DC 20006
202-835-7525

Complete if Known

(Use as many sheets as necessary)

of 2

Application Number	Not Yet Assigned
Filing Date	January 12, 2005
First Named Inventor	Laiho
Art Unit	Not Yet Assigned
Examiner Name	Not Yet Assigned
Attorney Docket Number	37488.01300US

[illegible]

FOREIGN PATENT DOCUMENTS						
Examiner Initials*	Cite No. ¹	Foreign Patent Document	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages Or Relevant Figures Appear	T ⁶
		Country Code ³ -Number ⁴ -Kind Code ⁵ (if known)				
		EP 1 375 584 A	01/02/2004	Borealis Technology Oy		
		EP 1 146 078 A	10/17/2001	Borealis Technology Oy		

Date
Considered

This collection of information is required by 37 CFR 1.97 and 1.98. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 2 hours to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.

If you need assistance in completing the form, call 1-800-PTO-9199 (1-800-786-9199) and select option 2.

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

Substitute for form 1449/PTO INFORMATION DISCLOSURE STATEMENT BY APPLICANT <i>(Use as many sheets as necessary)</i>		Complete if Known	
		Application Number	Not Yet Assigned
		Filing Date	January 12, 2005
		First Named Inventor	Laiho
		Art Unit	Not Yet Assigned
		Examiner Name	Not Yet Assigned
Sheet 2	of 2	Attorney Docket Number	37488.01300US

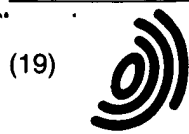
NON PATENT LITERATURE DOCUMENTS			
Examiner Initials*	Cite No. ¹	Include name of the author (in CAPITAL LETTERS), title of the article (when appropriate), title of the item (book, magazine, journal, serial, symposium, catalog, etc.), date, page(s), volume-issue number(s), publisher, city and/or country where published.	T ²
		International Search Report for PCT/ES2005/000221 dated July 29, 2005	
		Written Opinion for PCT/EP2005/000221 dated July 29, 2005	

Examiner Signature		Date Considered	
--------------------	--	-----------------	--

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant.

¹ Applicant's unique citation designation number (optional). ² Applicant is to place a check mark here if English language Translation is attached. This collection of information is required by 37 CFR 1.98. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 2 hours to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 (1-800-786-9199) and select option 2.



Europäisches Patentamt
European Patent Office
Office européen des brevets



(11)

EP 1 146 078 A1

(12)

EUROPEAN PATENT APPLICATION

(43) Date of publication:

17.10.2001 Bulletin 2001/42

(51) Int Cl.7: **C08L 23/06, C08L 23/04**

(21) Application number: **00108173.6**

(22) Date of filing: **13.04.2000**

(84) Designated Contracting States:

**AT BE CH CY DE DK ES FI FR GB GR IE IT LI LU
MC NL PT SE**

Designated Extension States:

AL LT LV MK RO SI

(72) Inventor: **Ahlstrand, Lars Erik**

44494 Ucklum (SE)

(74) Representative: **Kador & Partner**

Corneliusstrasse 15

80469 München (DE)

(71) Applicant: **Borealis Technology Oy**

06101 Porvoo (FI)

(54) **Polymer composition for pipes**

(57) HDPE compositions comprising a bimodal polymer and a nucleating agent, for production of pipes

having increased pressure resistance, and the use of such compositions in the production of pipes.

EP 1 146 078 A1

Description

[0001] This invention related to polymers for the production of pipes having increased pressure resistance.

[0002] Polyethylene pipe materials are often classified according to the design stress rating (ISO/DIS 12162.2). This is the circumferential stress the pipe is designed to withstand for at least 50 years without failure. The design stress rating is determined at different temperatures in terms of Minimum Required Strength (MRS) according to ISO/TR 9080. Thus, MRS8.0 means that the pipe can withstand an internal pressure of 8.0 MPa at 20°C for 50 years. Similarly, MRS10.0 means that the pipe can withstand a pressure of 10.0 MPa at the conditions above.

[0003] Pipes fulfilling the MRS8.0 requirements are typically made of either unimodal or bimodal ethylene polymers.

The pipes fulfilling the MRS 10.0 requirements are typically made of bimodal ethylene polymers. The corresponding polyethylene materials are often referred to as PE80 and PE100 materials, respectively.

[0004] Bimodal ethylene polymers have different densities and molecular weights depending on the intended use of the polymer. Thus, a bimodal ethylene polymer often used in pressure pipes comprises a bimodal ethylene polymer and a carbon black additive, having a density of about 955-961 kg/m³ and an MFR₅ of about 0.3-0.9 g/10 min. Another bimodal ethylene polymer used in pipe manufacture has a density of about 937-943 kg/m³ and an MFR₅ of about 0.5-1.0 g/10 min.

[0005] Bimodal PE100 materials have excellent properties compared to unimodal materials due to a high concentration of the tie chains which connect crystal lamellae. However, in natural bimodal materials the crystallites formed are large and the boundary layers between crystallites, consisting of segregated amorphous material, are relatively wide. Under these circumstances fewer tie chains will connect different crystallites.

[0006] It has been known to use nucleating agents to increase the crystallinity and decrease the crystal size of polypropylene. Polyethylene, however, has been known to have a much higher crystallization rate than polypropylene and nucleating agents have not been effective in it.

[0007] "PE100 Resins for Pipe Applications: Continuing the Development into the 21st Century" (Scheirs et al, TRIP Vol 4, No. 12, 1996) provides a summary of different PE100 grades on the market. It stresses the importance of the molecular structure, in specific molecular weight distribution and comonomer distribution, of the material.

[0008] EP-A-739937 discloses a pipe made of bimodal ethylene polymer, having a specific stress cracking resistance and impact strength. The material also has a specific stiffness and MFR.

[0009] The present invention is based on the surprising finding that a small amount of a nucleating agent in bimodal ethylene polymer composition significantly increases the pressure resistance of a pipe made from the polymer composition. While an increase of the pressure resistance has been observed with unimodal ethylene polymers also, the effect on bimodal compositions is dramatically and unexpectedly stronger. Typically, the nucleating agent also produces a reduction in the flexural modulus of the polymer.

[0010] According to the present invention a polymer composition for producing pipes with increased pressure resistance, comprises a bimodal polymer formed of:

- i) a low molecular weight ethylene polymer or copolymer and
- ii) a high molecular weight ethylene polymer or copolymer

and a nucleating agent, the alpha-olefin of the ethylene alpha-olefin copolymer preferably being selected from propylene, 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene, 1-octene, 1-decene and cyclic olefins, and constituting between 0.5 and 10 weight % of the copolymer. A carbon-black additive may also be present in the composition. The low molecular weight component (i) suitably comprises 30-70% by weight of the bimodal polymer.

[0011] In addition to the constituents mentioned above, the composition may also contain minor amounts of other components, such as prepolymer, carrier resins of masterbatches or similar, as disclosed in, e.g. WO-A-96/18677. The amount of such components should not exceed 5% of weight of the composition.

[0012] The bimodal polymer may be produced by blending the low and high molecular weight components in an extruder, or in a single- or multi-step polymerization process.

[0013] The nucleating agent may be a pigment or an additive which acts as a nucleus for a polyethylene crystal. Examples of such nucleating agents are the α - and β -phthalocyanine blue pigments and the phthalocyanine green pigment.

[0014] The bimodal ethylene polymer typically has a density of 930-965 kg/m³ and a melt index MFR₅ measured at 190°C under 5 kg load of 0.1-1.2 g/10 min, preferably of 0.15-1.0 g/10 min. It comprises preferably 40-55% and more preferably 43-48% by weight of low molecular weight component (i) and preferably 60-45% and more preferably 57-52% by weight of high molecular weight component (ii). It also preferably has a number average molecular weight M_n between about 8000 and 15000 g/mol, a weight average molecular weight M_w between about 180000 and 330000 g/mol, a polydispersity index M_w/M_n between about 20 and 40, and a content of units derived from the alpha-olefin comonomer between about 0.4 and 3.5% by mole.

[0015] The low molecular weight polymer (i) preferably has a weight average molecular weight of about 5000-50000 g/mol and a melt index MFR₂ measured under 2.16 kg load between about 50 and 2000 g/10 min. Preferably it is a homopolymer of ethylene, containing less than 2%, preferably less than 1%, more preferably less than 0.5% and most preferably less than 0.2% by mole units derived from higher alpha-olefin comonomers. Thus, the density of the low molecular weight ethylene polymer (i) should be 960-980 kg/m³, preferably 965-980 kg/m³ and more preferably between 970-980 kg/m³.

[0016] The high molecular weight polymer (ii) typically has a weight average molecular weight of about 300000-1000000 g/mol. Moreover, it is preferably a copolymer of ethylene and a higher alpha-olefin, the content of the alpha-olefin comonomer units being about 0.7-7.0% by mole. In particular, the molecular weight of the high molecular weight polymer (ii) should be such that when the low molecular weight ethylene polymer (i) has the melt index and density specified above, the bimodal polymer has the melt index and density as specified above.

[0017] The polymers (i) and (ii) may be produced using any known method where ethylene is polymerized in the presence of a Ziegler or a single site catalyst. Thus, they may be produced by a solution polymerization process, a slurry polymerization process or a gas phase polymerization process. The bimodal polymer may, moreover, be produced in a multistage process, such as that disclosed in EP-B-517868 or WO-A-96/18662. Typically, the polymerization takes place in the presence of a Ziegler catalyst, such as disclosed in EP-A-688794 and EP-A-949274. It is also possible to use a single site catalyst, such as that disclosed in FI-A-960437.

[0018] Preferably, the low molecular weight ethylene polymer (i) is produced in one stage of a multistage polymerization process and the high molecular weight ethylene polymer in another stage of the process. In particular, the low molecular weight ethylene polymer may be produced in a continuously operating loop reactor where ethylene is polymerized in the presence of a polymerization catalyst and a chain transfer agent, such as hydrogen. An inert aliphatic hydrocarbon, like isobutane or propane is used as a diluent. Preferably no, or only traces of a, higher alpha-olefin comonomer is present.

[0019] The hydrogen concentration should be selected so that the low molecular weight ethylene polymer (i) has the desired MFR. Typically, the molar ratio of hydrogen to ethylene is then between 0.1 and 1.0 mol/mol, preferably between 0.2 and 0.8 mol/mol. It is advantageous to operate the loop reactor using propane diluent in so called supercritical conditions, where the operating temperature exceeds the critical temperature of the reaction mixture and the operating pressure exceeds the critical pressure of the reaction mixture. A suitable range of temperature is then from 90 to 110°C and a suitable range of pressure is from 50 to 80 bar.

[0020] The slurry is intermittently or continuously removed from the loop reactor to a separation unit, where the hydrocarbons and especially the chain transfer agent are separated from the polymer. The polymer containing the active catalyst is introduced into a gas phase reactor, where the polymerization proceeds in the presence of additional ethylene, alpha-olefin comonomer and optionally, chain transfer agent to produce the high molecular weight ethylene polymer (ii). The polymer is intermittently or continuously withdrawn from the gas phase reactor and the remaining hydrocarbons are separated from the polymer. The polymer collected from the gas phase reactor is the ethylene polymer (1).

[0021] The conditions in the gas phase reactor are selected so that the bimodal polymer has the desired properties. Typically, the temperature in the reactor is between 70 and 100°C and the pressure is between 10 and 40 bar. The hydrogen to ethylene molar ratio ranges from 0.001 to 0.1 mol/mol and the alpha-olefin comonomer to ethylene molar ratio ranges from 0.05 to 0.5 mol/mol.

[0022] The nucleating agent may be any compound or mixture of compounds capable of nucleating the crystallization, such as a pigment having a nucleating effect or an additive used only for nucleating purposes. Examples of the first category of compounds are phthalocyanine blue or green pigments (e.g. PB15:1, PB15:3, PG7), isoindolinone and isoindoline pigments (e.g. PY109, PY110, PO61), benzimidazolone pigments (e.g. PO62, PO72), quinacridone pigments (e.g. PY19), benzimidazolone pigments (e.g. PY180, PY181), quinophtalone pigments (e.g. PY138), chinacridone pigments (e.g. Pigment Violet PV19) and azoheterocyclus pigments (e.g. PO64). Examples of the second category of compounds are dibenzylidene sorbitol derivatives and dibenzylidene xylitol derivatives.

[0023] The nucleating agent may also be a polymeric additive, such as a polymer of vinylcyclohexane or 3-methyl-1-butene. In such case, the polymeric additive, which preferably has a melting point above 200°C, may be blended into the bimodal polymer by conventional means in an extruder, or it may be prepolymerized on the catalyst as disclosed e.g. in WO 99/24478.

[0024] A characteristic feature of the invention is that a low amount of nucleating agent is needed to achieve the desired effect, usually significantly less than used previously in the art. This results in savings in raw material costs. Moreover, because a smaller amount of the additive is needed, there are less likely to be problems related to plate-out of the additive. The exact amount of the nucleating agent depends on which compound is used as the nucleating agent. The composition usually contains from about 1 to about 1500 ppm, preferably from 10 to 1000 ppm by weight of the nucleating agent.

[0025] Compositions according to the invention may also contain other additives known in the art, for instance sta-

bilizers such as hindered phenols, phosphates, phosphites and phosphonites, pigments such as carbon black, ultramarine blue and titanium dioxide, additives such as clay, talc, calcium carbonate, calcium stearate, and zinc stearate, UV absorbers, antistatic additives like those sold under trade name Lankrostat, and UV-stabilizers which may be hindered amines such as that sold under trade name Tinuvin 622.

[0026] According to one preferred embodiment of the invention, the bimodal polymer has a density of 943-953 kg/m³ and a melt index MFR₅ of 0.2-0.6 g/10 min. It comprises 45-55%, preferably 47-52% of low molecular weight homopolymer component (i) having MFR₂ of 300-700 g/10 min and 55-45%, preferably 53-48% by weight of high molecular weight component (ii). Additionally, it may contain 0-5% of other ethylene polymers having MFR₂ of 0.2-50 g/10 min and density of 920-980 kg/m³. The composition comprises 40-800 ppm of phthalocyanine blue as the nucleating agent. Further, the composition may contain 0-1000 ppm titanium dioxide, 0-5000 ppm ultramarine blue, 100-2000 ppm antioxidant (like Irganox 1010), 0-2000 ppm process stabilizer (like Irgafos 168), 0-3000 ppm calcium stearate or zinc stearate and 0-5000 ppm UV-stabilizer (like Tinuvin 622).

[0027] According to another preferred embodiment of the invention, the bimodal polymer has a density of 937-941 kg/m³ and a melt index MFR₅ of 0.7-1.1 g/10 min. It comprises 41-47%, preferably 42-46% of low molecular weight homopolymer component (i) having MFR₂ of 200- 500 g/10 min and 53-59 %, preferably 54-58 % by weight of high molecular weight component (ii). Additionally, it may contain 0-5% of other ethylene polymers having MFR₂ of 0.2-50 g/10 min and density of 920-980 kg/m³. The composition comprises 40-800 ppm of phthalocyanine blue as the nucleating agent. Further, the composition may contain 0-1000 ppm titanium dioxide, 0-5000 ppm ultramarine blue, 100-2000 ppm antioxidant (like Irganox 1010), 0-2000 ppm process stabilizer (like Irgafos 168), 0-3000 ppm calcium stearate or zinc stearate and 0-5000 ppm UV-stabilizer (like Tinuvin 622).

[0028] The classification of a composition comprising the bimodal polymer and additives and pigments, but not comprising the nucleating agent, can, by adding the nucleating agent into the composition as prescribed by the invention, be increased from a value of MRS 10.0 to a value of MRS 11.2, which connotes a major improvement in pressure resistance of pipe made from the composition.

[0029] The invention is illustrated by the following Examples, in which MFR is measured according to ISO 1133 at 190°C. The load has been indicated as a subscript, i.e. MFR₂ denotes that the measurement has been carried out under a load of 2.16 kg.

[0030] The pressure test values originate from slow crack propagation resistance test, performed according to ISO 1167. The resulting figure indicates how many hours the pipe can withstand a certain pressure at a certain temperature without a failure. The temperature and pressure are indicated as test parameters, e.g. 20°C/12.4 MPa indicates that the test was performed at 20°C temperature with 12.4 MPa pressure within the pipe. Briefly, the test procedure is as follows.

[0031] Pipes with 32 mm diameter and a thickness of 3 mm, internally filled with water, are mounted in a water bath and connected to a device which allows the internal water pressure to be adjusted and controlled within a range of +2 to - 1%. The temperature of the water bath can be selected and is kept constant to within a mean of +/- 1°C. The time to pipe failure is registered automatically.

Example 1

[0032] The base polymer is a natural bimodal material produced in two cascaded CSTR slurry reactors using a polymerization catalyst containing Mg and Ti as active ingredients. In the first reactor diluent, ethylene and hydrogen were added together with the catalyst and triethylaluminum cocatalyst, so that an ethylene homopolymer with MFR₂ of 500 g/10 min was produced. The slurry was withdrawn from the first reactor, excess hydrogen and ethylene were removed and the polymerization was continued in the second reactor by adding ethylene, hydrogen and 1-butene comonomer. The slurry was withdrawn from the reactor, the hydrocarbons were removed and the polymer was compounded in an extruder and pelletized. The MFR₅ of the final polymer was 0.4 g/10 min and the density 948.

[0033] In runs 1-5 the polymerization catalyst was non silica-based and the production split (between first and second reactors) was 52/48. In runs 6-9 the polymerization catalyst was silica-based and the production split was 48/52.

[0034] Various pigment formulations were then worked into the pelleted material on a Buss Kneader 100-11D, and the thoroughly mixed compositions were extruded into 32 mm diameter pipes of 3 mm wall thickness which were subjected to the above described ISO 1167 test. Of the various components of these formulations, the titania, the ultramarine and the yellow pigment "PY93" are devoid of nucleating effect. The results are shown in Table 1 below.

TABLE 1

Run	Temp/Stress	Pigment Formulation (ppm) (total composition basis)	Hours to Failure
1	80°/5.7 MPa	None	370
2	80°/5.7 MPa	PY93*: 1500 ppm TiO ₂ : 600 ppm	91
3	80°/5.7 MPa	PB15.1**: 800 Ultramarine: 2500 TiO ₂ : 900	1280
4	80°/5.7 MPa	PB15.3**: 1775 TiO ₂ : 945	1510
5	80°/5.7 MPa	PB29***: 3800 TiO ₂ : 370 PY93: 50	130
6	20°/12.4 MPa	None	83†

7	80°/5.7 MPa	None	460††
8	20°/12.4 MPa	PB15.1: 375 PB29: 1275 TiO ₂ : 405	7873†
9	80°/5.7 MPa	PB15.1: 375 PB29: 1275 TiO ₂ : 405	>17800 (running)

* o-benzotoluidide, 3,3''-[(2-chloro-5-methyl-p-phenylene) bis (immocarbonylacetonilideneazo)] bis [3',4-dichloro-(8Cl)], a yellow pigment.

** denote $\alpha(.1)$ - and $\beta(.3)$ - Cu phthalocyanine blue.

*** ultramarine

† mean of two tests.

†† mean of five tests.

Example 2

[0035] The base polymer is a natural bimodal material produced in a process comprising a cascade of a loop reactor and a gas phase reactor, in presence of a silica based catalyst. In the loop reactor diluent, ethylene and hydrogen were added together with the catalyst and triethylaluminum cocatalyst, so that an ethylene homopolymer with MFR₂ of 350 g/10 min was produced. The slurry was withdrawn from the first reactor, hydrocarbons were removed and the polymerization was continued in the gas phase reactor by adding ethylene, hydrogen and 1-butene comonomer. The polymer was withdrawn from the reactor, the hydrocarbons were removed and the polymer was compounded in an extruder and pelletized. The MFR₅ of the final polymer was 0.93 g/10 min and the density 940. The production split was 44% in the loop reactor, 56% in the gas phase reactor.

[0036] Test batches were prepared and subjected to test as in Example 1. The results are shown in Table 2 below.

TABLE 2

Temp/Stress	Pigment Formulation (ppm) (Total Composition Basis)	Hours to Failure
20°/10.0 MPa	None (3000 ppm Tinuvin 622 added as UV stabilizer)	208
80°/4.6 MPa	None (3000 ppm Tinuvin 622 added as UV stabilizer)	0.9
20°/10.0 MPa	PB29:1200 PB15.1: 660 PG7*:230 TiO ₂ :4400 (plus 2000 ppm Tinuvin 622 as UV stabilizer)	Test concluded at 3000
80°/4.6 MPa	PB29:1200 PB15.1:660 PG7:230 TiO ₂ :4400 (plus 2000 ppm Tinuvin 622 as UV stabilizer)	3032

* phthalocyanine green

Example 3 (comparative)

[0037] The base polymer is a natural unimodal material produced in a gas phase process in presence of a silica based chromium catalyst. Ethylene, 1-butene and polymerization catalyst were introduced into the reactor operated in such conditions that a polymer resin having a MFR₅ of 0.9 g/10 min and the density 939 kg/m³ was obtained.

[0038] Test batches were prepared and subjected to test as in the previous Examples.
The results are shown in Table 3 below.

TABLE 3

Temp/Stress	Pigment Formulation (ppm) (Total Composition Basis)	Hours to Failure
20°/11.0 MPa	PY93 : 990	230
20°/11.0 MPa	PB29:1200 PB15.1:660 PG7:230 TiO ₂ : 4400	400

[0039] It will be observed that when the base PE resin is unimodal the effect of the phthalocyanine blue nucleating agent is not significantly greater than that of the non-nucleating yellow pigment, and is significantly lower than its effect at comparable conditions when (Table 2) the base PE resin is bimodal.

Claims

1. A polymer composition comprising an ethylene homopolymer or an ethylene alpha-olefin copolymer for producing pipes with increased pressure resistance, wherein the polymer is a bimodal polymer comprising
 - i) a low molecular weight ethylene polymer or copolymer and
 - ii) a high molecular weight ethylene polymer or copolymer
 and a nucleating agent.
2. A polymer composition according to claim 1, **characterized in that** the alpha-olefin of the ethylene alpha-olefin copolymer is selected from the group of propylene, 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene, 1-octene, 1-decene and cyclic olefins.
3. A polymer composition according to claims 1 or claim 2, **characterized in that** the alpha-olefin comonomer content is between 0.5 and 10 weight %.
4. A polymer composition according to any of claims 1 to 3, **characterized in that** a carbon-black additive is present.
5. A polymer composition according to any preceding claim, **characterized in that** the density is between 930 and 965 kg/m³.
6. A polymer composition according to any preceding claim, **characterized in that** the melt index MFR5 measured at 190°C under 5 kg load is between 0.1 and 1.2 g/10 min.
7. A polymer composition according to any preceding claim, **characterized in that** the polymer composition comprises 30-70% of said low molecular weight ethylene polymer or copolymer and 70-30% of said high molecular weight ethylene polymer or copolymer.
8. A polymer composition according to any preceding claim, **characterized in that** the number average molecular

weight Mn is between 8000 and 150000 g/mol.

9. A polymer composition according to any preceding claim, **characterized in that** the weight average molecular weight Mw is between 180000 and 330000 g/mol.

10. A polymer composition according to any preceding claim, **characterized in that** the polydispersity index Mw/Mn is between 20 and 40.

11. A polymer composition according to any preceding claim, **characterized in that** the low molecular weight ethylene polymer or copolymer has a weight average molecular weight of between 5000 and 50000 g/mol.

12. A polymer composition according to any preceding claim, **characterized in that** the low molecular weight ethylene polymer or copolymer has a melt index MFR2 measured under 2.16 kg load of between 50 and 2000 g/10min.

13. A polymer composition according to any preceding claim, **characterized in that** the low molecular weight ethylene copolymer has a comonomer content of less than 2 mol %.

14. A polymer composition according to any preceding claim, **characterized in that** the low molecular weight ethylene polymer or copolymer has a density between 960 and 980 kg/m³.

15. A polymer composition according to any preceding claim, **characterized in that** the high molecular weight ethylene polymer or copolymer has a weight average molecular weight of between 300000 and 1000000 g/mol.

16. A polymer composition according to any preceding claim, **characterized in that** the high molecular weight ethylene copolymer has a comonomer content of 0.7 to 7.0 mol %.

17. A polymer composition according to any preceding claim, **characterized in that** the nucleating agent is a pigment.

18. A polymer composition according to any preceding claim, **characterized in that** the nucleating agent is a phthalocyanine blue or green pigment.

19. A polymer composition according to any of claims 1 to 16, **characterized in that** the nucleating agent is a dibenzylidene sorbitol derivative.

20. A polymer composition according to any of claims 1 to 16, **characterized in that** the nucleating agent is a dibenzylidene xylitol derivative.

21. A polymer composition according to any of claims 1 to 16, **characterized in that** the nucleating agent is a polymeric additive.

22. A polymer composition according to any preceding claim, **characterized in that** the nucleating agent is present in an amount of between 1 and 1500 ppm.

23. A polymer composition comprising an ethylene homopolymer or an ethylene alpha-olefin copolymer for producing pipes with increased pressure resistance, wherein the polymer is a bimodal polymer comprising

- i) a low molecular weight ethylene polymer or copolymer and
- ii) a high molecular weight ethylene polymer or copolymer

and a nucleating agent, whereby the pressure resistance MRS according to ISO/TR 9080 is at least 10% higher than for the same polymer composition not comprising a nucleating agent.

24. Use of a polymer composition comprising an ethylene homopolymer or an ethylene alpha-olefin copolymer, whereby the polymer is a bimodal polymer and comprises

- i) a low molecular weight ethylene polymer or copolymer and
- ii) a high molecular weight ethylene polymer or copolymer and a nucleating agent for producing pipes with increased pressure resistance.



European Patent
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EUROPEAN SEARCH REPORT

Application Number
EP 00 10 8173

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (IntCl7)
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The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 4 July 2000	Examiner Bergmans, K
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons ----- & : member of the same patent family, corresponding document	

**ANNEX TO THE EUROPEAN SEARCH REPORT
ON EUROPEAN PATENT APPLICATION NO.**

EP 00 10 8173

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report.
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(54) **Breathable Films**

(57) The present invention concerns a composition for preparing breathable films. The composition comprises a bimodal polyethylene composition, a particulate filler, and optionally, an olefin-based polymer. The bimodal polyethylene composition has a melt flow rate MFR₂ of 0.1 to 4 g/10 min and a density of 918 to 935 kg/m³. The olefin-based polymer can be, e.g., polypropylene.

The films prepared from the composition have a very high water vapour transmission rate, exceeding 3000 g/m²/24 h. The composition can be prepared to thin films having a low basis weight, of 25 g/m² or less. The films have no pinholes.

Description**Background of the Invention****Field of the Invention**

[0001] The present invention concerns breathable films prepared from linear low-density polyethylene compositions. In addition, the present invention concerns bimodal linear low-density polyethylene compositions used for preparing breathable films. In particular, the present invention relates to breathable films having an improved mechanical strength.

Description of Related Art

[0002] It is known in the art to prepare breathable films by blending thermoplastic polymers with fillers and stretching the films so, that voids are formed adjacent to the filler particles.

[0003] WO-A-01/79343 discloses a microporous thermoplastic film having an improved impact strength and high moisture vapour transmission rate. The film is prepared from a blend containing 40 to 60 % calcium carbonate, 30 to 40 % linear low density PE and 1 to 10 % low density PE. The film is then incrementally stretched to provide the microporous film.

[0004] WO-A-99/32164 discloses an absorbent article with a topsheet, backsheet and an absorbent layer between the two. The backsheet comprises a microporous polymer film containing 30 to 60 % polyolefin and 40 to 80 % calcium carbonate. After the film is cast, it is drawn to form the microporous holes around the calcium carbonate filler. Polyethylene was used in the example.

[0005] WO-A-99/14262 discloses a breathable film made of a composition containing a first ethylene polymer, having a density lower than 890 kg/m³, a second ethylene polymer having a density above 900 kg/m³ and at least 35 % of a filler. The ratio between the first ethylene polymer and the second ethylene polymer is 25/75 - 75/25. The film was stretched to make it porous. The examples showed that metallocene based PE was used both as the first ethylene polymer and the second ethylene polymer.

[0006] While the above documents disclose different breathable films and compositions for preparing them, there still remains a need for films having a high water vapour transmission rate combined with good mechanical properties and good processability.

Summary of the Invention

[0007] It is an object of the present invention to provide breathable films having good mechanical properties and good processability.

[0008] These and other objects, together with the advantages thereof over known processes and products, which shall become apparent from the specification, which follows, are accomplished by the invention as hereinafter described and claimed.

[0009] The present invention is based on the provision of compositions comprising:

- (i) 20 - 50 %, based on the weight of the total composition, a bimodal polyethylene composition comprising
- (ii) a first (low molecular weight) component with a melt flow rate MFR₂ of 50 to 500 g/10 min, preferably of 100 to 400 g/10 min and a density of 940 to 975 kg/m³, preferably 945 to 975 kg/m³, the first component being present in the bimodal polyethylene composition in an amount of 37 to 48 % by weight,
- (iii) at least one other component having a higher molecular weight (or a lower melt flow rate) and a lower density than the said first component, the second component being present in the bimodal polyethylene composition in an amount of 52 to 63 % by weight, so that the said bimodal polyethylene composition has a melt flow rate MFR₂ in the range of 0.1 to 4.0 g/10 min, preferably 0.1 to 0.8 g/10 min, MFR₂₁ in the range of 15 to 200 g/10 min, preferably 15 to 70 g/10 min and a density of 918 to 935 kg/m³,
- (iv) 40 - 70 %, based on the weight of the total composition, a particulate filler, and
- (v) 0 - 30 %, based on the weight of the total composition, another olefin-based polymer

[0010] More specifically, the process according to the present invention is mainly characterised by what is stated in the characterising part of claim 1.

[0011] Additionally, the present invention provides a process for producing the polymer composition. First, the said bimodal polyethylene composition is produced in situ by polymerising or copolymerising ethylene in a reactor cascade formed by at least a first reactor and a second reactor in the presence of a polymerisation catalyst. The polymerisation catalyst has been prepared by supporting a magnesium compound, an aluminium compound and a titanium compound

on a particulate support. Second, the bimodal composition is blended with the particulate filler and optionally, the other olefin based polymer.

[0012] One more aspect of the present invention is to provide breathable, microporous films having improved properties.

[0013] A further aspect of the invention is to provide the use of the above-mentioned composition for breathable films.

[0014] Still one more aspect of the invention is to provide a process for preparing breathable films.

[0015] Next, the invention will be more closely examined with the aid of the following detailed description and examples.

Detailed Description of the Invention

Definitions

[0016] For the purpose of the present invention, "slurry reactor" designates any reactor operating in slurry, in which reactor the polymer forms in particulate form. As examples of suitable reactors can be mentioned a continuous stirred tank reactor, a batch-wise operating stirred tank reactor or a loop reactor. According to a preferred embodiment the slurry reactor comprises a loop reactor.

[0017] By "gas phase reactor" is meant any mechanically mixed or fluidised bed reactor. Preferably the gas phase reactor comprises a fluidised bed reactor with gas velocities of at least 0.2 m/sec, which may further have a mechanical agitation.

[0018] By "melt flow rate" or abbreviated "MFR" is meant the weight of a polymer extruded through a standard cylindrical die at a standard temperature (190 °C for polyethylene) in a laboratory rheometer carrying a standard piston and load. MFR is a measure of the melt viscosity of a polymer and hence also of its molecular weight. The abbreviation "MFR" is generally provided with a numerical subscript indicating the load of the piston in the test. Thus, e.g., MFR₂ designates a 2.16 kg load. MFR can be determined using, e.g., by one of the following tests: ISO 1133 C4, ASTM D 1238 and DIN 53735.

The composition

[0019] The present invention provides a composition for making breathable films having a high rate of water vapour transmission (WVTR), the composition comprising:

- (i) 20 - 50 %, based on the weight of the total composition, a bimodal polyethylene composition comprising
- (ii) a first (low molecular weight) component with a melt flow rate MFR₂ of 50 to 500 g/10 min, preferably of 100 to 400 g/10 min and a density of 940 to 975 kg/m³, preferably 945 to 975 kg/m³, the first component being present in the bimodal polyethylene composition in an amount of 37 to 48 % by weight,
- (iii) at least one other component having a higher molecular weight (or a lower melt flow rate) and a lower density than the said first component, the second component being present in the bimodal polyethylene composition in an amount of 52 to 63 % by weight, so that the said bimodal polyethylene composition has a melt flow rate MFR₂ in the range of 0.1 to 4.0 g/10 min, preferably 0.1 to 0.8 g/10 min, MFR₂₁ in the range of 15 to 200 g/10 min, preferably 15 to 70 g/10 min and a density of 918 to 935 kg/m³,
- (iv) 40 - 70 %, based on the weight of the total composition, a particulate filler, and
- (v) 0 - 30 %, based on the weight of the total composition, another olefin-based polymer

Bimodal polyethylene composition

[0020] The use of bimodal polyethylene component gives the compositions of the present invention a high mechanical strength. It also gives the compositions a good processability and allows the preparation of thin films having a low basis weight. Very high water vapour transmission rates can be reached, with no pinholes in the film.

[0021] As referred to above, the bimodal polyethylene composition comprises 20 - 50 % of the composition, based on the total weight of the composition. The bimodal polyethylene composition further comprises of 37 - 48 % of a low molecular weight component and 52 - 63 % of a high molecular weight component, based on the weight of the bimodal polyethylene composition.

[0022] The low molecular weight component helps to improve the processability of the composition. It has an MFR₂ of about 50 to 500 g/10 min, preferably 100 to 400 g/10 min. It may contain a C₄- C₁₀ alpha-olefin comonomer so that it has a density of about 940 kg/m³ or higher, preferably of about 945 kg/m³ or higher, but it may also be a homopolymer having a density of about 975 kg/m³.

[0023] The high molecular weight component gives the mechanical properties to the composition. It is a copolymer

of ethylene and a $C_4 - C_{10}$ alpha-olefin, and it has a higher molecular weight and a higher content of comonomer than the low molecular weight component. It has such molecular weight and comonomer content that at given properties of the low molecular weight component and at a given split of the components, the bimodal polyethylene composition has the desired melt index and density.

[0024] According to one preferred embodiment of the invention, the low molecular weight component is a copolymer of ethylene and a $C_4 - C_{10}$ alpha-olefin, having a melt flow rate MFR_2 of 50 to 500 g/10 min, preferably of 100 to 400 g/10 min and a density of 940 to 955 kg/m³, preferably 945 to 953 kg/m³. The bimodal polyethylene composition has a melt flow rate MFR_2 of 0.4 to 0.8 g/10 min, and a density of 918 to 925 kg/m³.

[0025] According to another preferred embodiment of the invention, the low molecular weight component is a copolymer of ethylene and a $C_4 - C_{10}$ alpha-olefin, having a melt flow rate MFR_2 of 100 to 500 g/10 min, preferably of 200 to 400 g/10 min and a density of 940 to 955 kg/m³, preferably 945 to 953 kg/m³. The bimodal polyethylene composition has a melt flow rate MFR_2 of 0.1 to 0.3 g/10 min, MFR_{21} of 15 to 35 g/10 min and a density of 918 to 925 kg/m³.

[0026] According to still another preferred embodiment of the invention, the low molecular weight component is a homopolymer of ethylene having a melt flow rate MFR_2 of 100 to 500 g/10 min, preferably of 200 to 400 g/10 min and a density of about 975 kg/m³. The bimodal polyethylene composition has a melt flow rate MFR_2 of 0.1 to 0.3 g/10 min, MFR_{21} of 15 to 35 g/10 min and a density of 925 to 935 kg/m³.

Particulate filler

[0027] The particulate filler is a solid material in the form of particles, which can be uniformly dispersed over the film. Advantageously, the particulate filler has an average particle size within the range of 0.1 to 10 μm , preferably 0.1 to 4 μm . Examples of such fillers are calcium carbonate, magnesium carbonate, barium carbonate, sodium carbonate, different clays, silica, alumina, barium sulphate, diatomaceous earth, magnesium sulphate, mica, carbon, calcium oxide, magnesium oxide etc. The filler particles may also be coated with a fatty acid to improve the flow properties of the particles. Calcium carbonate is an especially preferred particulate filler.

[0028] The particulate filler comprises 40 - 70 % of the total weight of the composition. It is the present understanding that when the composition is extruded to a film and the film is stretched, micropores are formed adjacent to the filler particles. These micropores allow the passage of gases and vapours through the film. On the other hand, the micropores are small enough to prevent the passage of liquids through the film.

Olefin-based polymer

[0029] The olefin-based polymer, which may be present in the compositions of the present invention, may be a homo- or copolymer of ethylene, propylene, 1-butene, 4-methyl-1-pentene etc, which is different from the bimodal polyethylene composition referred to above. Thus, it has been found that high-impact propylene copolymers are suitable to be used in the present invention. Another useful polymer which may be used is a bimodal linear low density polyethylene used for extrusion coating, having a melt index MFR_2 of about 10 g/10 min and a density of about 927 kg/m³.

Process for making the composition

Polymerisation catalyst

[0030] The polymerisation catalyst contains a magnesium compound, an aluminium compound and a titanium compound supported on a particulate support.

[0031] The particulate support can be an inorganic oxide support, such as silica, alumina, titania, silica-alumina and silica-titania. Preferably, the support is silica.

[0032] The average particle size of the silica support can be typically from 10 to 100 μm . However, it has turned out that special advantages can be obtained if the support has an average particle size from 15 to 30 μm , preferably from 18 to 25 μm . Especially it has been found out that the average particle size of the polymer produced in the process of the invention is the same irrespective whether the catalyst is prepared on a 20 μm support or on a 40 μm support. In fact, the fraction of fine polymer particles has been found to be lower if a support having an average particle size of 20 μm is used. The reduction of the fine polymer reduces the risk of plugging and thus contributes to a stable process operation. This, on the other hand, helps to produce polymer films with a good homogeneity.

[0033] The magnesium compound is a reaction product of a magnesium dialkyl and an alcohol. The alcohol is a linear or branched aliphatic monoalcohol. Preferably, the alcohol has from 6 to 16 carbon atoms. Branched alcohols are especially preferred. 2-ethyl-1-hexanol is one example of the preferred alcohols. The magnesium dialkyl may be any compound of magnesium bonding to two alkyl groups, which may be the same or different. Butyl-octyl magnesium is one example of the preferred magnesium dialkyls.

[0034] The aluminium compound is chlorine containing aluminium alkyl. Especially preferred compounds are aluminium alkyl dichlorides and aluminium alkyl sesquichlorides.

[0035] The titanium compound is a halogen containing titanium compound, preferably chlorine containing titanium compound. Especially preferred titanium compound is titanium tetrachloride.

[0036] The catalyst can be prepared by sequentially contacting the carrier with the above mentioned compounds, as described in EP-A-688794. Alternatively, it can be prepared by first preparing a solution from the components and then contacting the solution with a carrier, as described in WO-A-01/55230.

[0037] The above mentioned solid catalyst component is contacted with a aluminium alkyl cocatalyst, which preferably is an aluminium trialkyl compound, after which it can be used in polymerisation. The contacting of the solid catalyst component and the aluminium alkyl cocatalyst can either be conducted prior to introducing the catalyst into the polymerisation reactor, or it can be conducted by introducing the two components separately into the polymerisation reactor.

Polymerisation process

[0038] To produce the polymer compositions according to the invention, ethylene is polymerised in the presence of a polymerisation catalyst at elevated temperature and pressure. Polymerisation is carried out in a series of polymerisation reactors selected from the group of slurry and gas phase reactors. In the most preferred embodiment, the reactor system comprises one loop reactor (referred to in the subsequent text as "the first reactor") and one gas phase reactor (referred to in the subsequent text as "the second reactor"), in that order.

[0039] However, it should be understood that the reactor system can comprise other reactors in addition to the first and the second reactor. Thus, it is possible to include reactors, e.g. for prepolymerisation, or to divide either of the reactors in two or more reactors.

[0040] The high molecular weight portion and the low molecular weight portion of the product can be prepared in any order in the reactors. A separation stage is normally needed between the reactors to prevent the carryover of reactants from the first polymerisation stage into the second one. The first stage is typically carried out using an inert reaction medium.

[0041] The catalyst used in the polymerisation process can be a Ziegler-Natta or a metallocene catalyst. According to a preferred embodiment, a Ziegler-Natta catalyst is used. According to another preferred embodiment, no fresh catalyst is added to the second polymerisation stage.

[0042] In every polymerisation step it is possible to use also comonomers selected from the group of C_{3-18} olefins, preferably C_{4-10} olefins, such as 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene, 1-heptene, 1-octene, 1-nonene and 1-decene as well as mixtures thereof.

[0043] In addition to the actual polymerisation reactors used for producing the bimodal ethylene homo- or copolymer, the polymerisation reaction system can also include a number of additional reactors, such as prereactors. The prereactors include any reactor for prepolymerising the catalyst and for modifying the olefinic feed, if necessary. All reactors of the reactor system are preferably arranged in series (in a cascade).

[0044] According to a preferred embodiment of the invention, the polymerisation comprises the steps of:

- (i) subjecting ethylene, hydrogen and optionally comonomer(s) to a first polymerisation or copolymerisation reaction in the presence of the polymerisation catalyst in a first reaction zone in a loop reactor to produce a first reaction product having a low molecular weight with a melt flow rate MFR_2 of 50 to 500 g/10 min, preferably of 100 to 400 g/10 min and a density of 940 to 975 kg/m³, preferably 945 to 975 kg/m³,
- (ii) recovering the first polymerisation product from the first reaction zone,
- (iii) feeding the first polymerisation product to a second reaction zone or reactor,
- (iv) feeding additional ethylene, comonomers and optionally hydrogen to the second reaction zone,
- (v) subjecting the additional ethylene and additional comonomer(s) and optionally hydrogen to a second polymerisation reaction in the presence of the said polymerisation catalyst and the first polymerisation product,
- (vi) to produce a polymer composition comprising from 41 to 48 % by weight of the low molecular weight polymer produced in step (i), and from 59 to 52 % by weight of the high molecular weight component produced in step (v),
- (vii) the composition having a melt flow rate in the range MFR_2 of 0.1 to 4.0 g/10 min, preferably 0.1 to 0.8 g/10 min and a density of 918 to 935 kg/m³, and
- (viii) recovering the combined polymerisation product from the second reaction zone.

[0045] In the first step of the process, ethylene with the comonomer(s) is fed into the first polymerisation reactor. Along with these components is fed also hydrogen, which functions as a molecular weight regulator. The amount of hydrogen depends on the desired molecular weight of the polymer. The catalyst may be fed to the reactor together with the reagents or, preferably, in a separate stream, normally by flushing with a diluent.

[0046] The polymerisation medium typically comprises the monomer (i.e. ethylene) and/or a hydrocarbon, in partic-

ular, a light inert hydrocarbon, such as propane, isobutane, n-butane or isopentane. The fluid is either liquid or gaseous. In the case of a loop reactor, the fluid is liquid and the suspension of polymer is circulated continuously through the slurry reactor, whereby a suspension of polymer in particle form in a hydrocarbon medium or monomer will be produced.

[0047] The conditions of the loop reactor are selected so that 37 - 48 wt-%, preferably 39 - 47 wt-%, of the whole production is polymerised in the loop reactor(s). The temperature is in the range of 40 to 110 °C, preferably in the range of 70 to 100 °C. The reaction pressure is in the range of 25 to 100 bar, preferably 35 to 80 bar. The mole fraction of ethylene in the reaction mixture is typically of 4 to 10 %, preferably of 5 to 9 %. The ratio of the alpha-olefin comonomer to ethylene depends on the density of the polymer that is produced in the first stage; typically it is of 0 to 800 mol/kmol.

[0048] Hydrogen is also fed into the first reactor to control the molecular weight (or melt flow rate) of the polymer. The exact ratio of hydrogen to ethylene depends on the desired melt flow rate of the polymer to be produced; typically it is of 100 to 600 mol/kmol, preferably of 150 to 400 mol/kmol.

[0049] The polymerisation heat is removed by cooling the reactor with a cooling jacket. The residence time in the slurry reactor must be at least 10 minutes, preferably 40-80 min for obtaining a sufficient degree of polymerisation.

[0050] After the first reaction zone at least part of the volatile components of the reaction medium are evaporated. As a result of the evaporation, at least the major part of hydrogen is removed from the product stream. The product stream is then subjected to a second polymerisation stage in the gas phase reactor in the presence of additional ethylene to produce a high molecular weight polymer.

[0051] The second reactor is a gas phase reactor, wherein ethylene, comonomers and preferably hydrogen are polymerised in a gaseous reaction medium in the presence of the polymerisation catalyst.

[0052] The gas phase reactor can be an ordinary fluidised bed reactor, although other types of gas phase reactors can be used. In a fluidised bed reactor, the bed consists of the formed and growing polymer particles as well as still active catalyst that enters the reactor with the polymer stream. The bed is kept in a fluidised state by introducing gaseous components, for instance monomer and comonomer(s) from the bottom of the reactor on such a flow rate that the particles are supported but not entrained by the gas stream. The fluidising gas can contain also inert gases, like nitrogen and propane and also hydrogen as a molecular weight modifier. The fluidised bed gas phase reactor can be equipped with a mechanical mixer.

[0053] The gas phase reactor used can be operated in the temperature range of 50 to 115 °C, preferably between 60 and 100 °C and the reaction pressure between 10 and 40 bar and the partial pressure of ethylene between 2 and 20 bar, preferably between 3 and 8 bar.

[0054] The production split between the low molecular weight polymerisation reactor and the high molecular weight polymerisation reactor is (37 to 48 %): (63 to 52 %), based on the weight of the polymer composition. Preferably, 39 to 47 wt- of the ethylene copolymer is produced at conditions to provide a polymer having an MFR₂ of 50 to 500 g/10 min, preferably of 100 to 400 g/10 min and a density 940 to 975 kg/m³, preferably 945 to 975 kg/m³. Respectively, it is preferred that 53 to 61 % of the ethylene copolymer is produced at conditions to provide the high molecular weight polymer, having been produced in such conditions that the final polymer composition has an MFR₂ of 0.1 to 4.0 g/10 min, preferably 0.1 to 0.8 g/10 min, and a density of 918 to 925 kg/m³.

[0055] As mentioned above, the ratio of comonomer to ethylene in the second reactor is selected so that the final polymer composition has the desired density. A suitable range is 500 to 900 mol/kmol, preferably 500 to 800 mol/kmol.

[0056] In a similar fashion, the ratio of hydrogen to ethylene in the second reactor is selected so that the final polymer composition has the desired melt flow rate. A typical range is 1 to 30 mol/kmol, preferably 3 to 20 mol/kmol.

[0057] The present polymers and copolymers of ethylene can be blended and optionally compounded with additives and adjuvants conventionally used in the art. Thus, suitable additives include antistatic agents, flame retardants, light and heat stabilisers, pigments and processing aids.

Compounding

[0058] After the polymer is collected from the reactor and the hydrocarbon residues are removed therefrom, the polymer is compounded and extruded to pellets. In this process step, any extruder known in the art may be used. It is preferred, however, to use a twin screw extruder. It may be of a co-rotating type, such as those produced by Werner & Pfleiderer having a designation ZSK, e.g. ZSK 90 having a 90 mm screw diameter. Alternatively, it may be of a counter-rotating type, such as those produced by Japan Steel Works, having a designation JSW CIM-P, e.g. CIM90P, having a 90 mm screw diameter. It is especially preferred to use a counter-rotating twin screw extruder.

[0059] The particulate filler and optionally, the olefin-based polymer may be added to the bimodal polyethylene composition at this extrusion stage. It is possible, however, to mix the bimodal polyethylene composition with additives, and extrude it to pellets. These pellets are then introduced into a second extrusion stage, to which also the particulate filler and optionally, the olefin-based polymer, is introduced. The thus obtained compound may then be extruded directly into a film. However, it may also be extruded to pellets, which are collected and extruded to a film in a separate extrusion stage.

Films

[0060] The composition according to the present invention is used to prepare breathable films. The films may be produced either by blowing or casting. The polymers having a melt index at the lower end of the MFR range, having MFR₂ of 0.1 to 0.8 g/10 min, are suitable for film blowing. On the other hand, the polymers having a melt index at the higher end of the MFR range, having MFR₂ of 0.4 to 4.0 g/10 min, are suitable for making cast films.

[0061] After the film has been prepared, it shall be stretched. The purpose of stretching is to produce micropores adjacent to the filler particles, thus making the film breathable. The film shall be stretched from 3 to 10 times, preferably 4 to 7 times, its original length. This ratio between the length of the stretched film and the length of the original film is in the subsequent text referred to as the stretching ratio.

[0062] Surprisingly, the films of the present invention have a very high water vapour transmission rate. To achieve this high rate, it is advantageous to use a high fraction of filler particles (from 57 to 70 %) in the composition, preferably together with a high stretching ratio (from 5.5 to 7).

[0063] It appears that the high mechanical strength and the good processability of the bimodal polyethylene composition make it possible to use high stretching ratios. This allows to reach a very high water vapour transmission rate, higher than 3000 g/m²/24 h, in fact even higher than 4000 g/m²/24 h.

[0064] Alternatively and surprisingly, water vapour transmission rate higher than 3000 g/m²/24 h, or even higher than 4000 g/m²/24 h can be obtained by providing a composition comprising 25 to 40 % of the bimodal polyethylene composition, 50 to 57 % of the particulate filler and 5 to 20 % of a propylene polymer. When this composition is prepared into a film and stretched with a stretching ratio of 4 to 5.5, the resulting film has the high water vapour transmission rate referred to above.

[0065] The effect of the presence of the propylene polymer on the water vapour transmission rate is surprisingly strong. It was found that the rate could be increased by more than 100 % by adding the propylene polymer into the composition, compared to a similar composition where the propylene polymer was not present. The propylene polymers that can be used to increase the water vapour transmission rate include, propylene homopolymers, random copolymers of propylene with other olefins, especially ethylene, high impact propylene copolymers and propylene-ethylene rubbers. It is believed that polymers of other olefins, such as 1-butene or 4-methyl-1-pentene would have a similar effect.

[0066] Also, thin films having a low basis weight can be obtained without pinholes. Thus, the films of the present invention have can have a thickness of 25 µm or less, even 20 µm or less, and they can have a basis weight of 25 g/m² or less, even 20 g/m² or less. This makes it possible to prepare the films from a smaller amount of polymer, thus allowing to save in raw material costs.

[0067] One more surprising advantage of the use of the bimodal polyethylene composition as a base polymer in the composition is the reduction of the amount of scrap material when producing the films and the compositions, compared with the situation when a unimodal polyethylene is used as a base polymer in the composition. It appears that the use of the bimodal polyethylene composition gives a good homogeneity of the composition, and therefore the amount of waste is substantially reduced. This improves the economy of the film preparation process.

Description of Analytical Methods

Tensile strength

[0068] The experiment is performed according to ISO 1184 method. The specimen is extended along its major axis at a constant speed. Normal 50 mm could be used as a distance between grips (gauge length) in film tensile testing. 125 mm gauge length would be required for tensile modulus measurement so this was not possible for 100 mm cast film in transverse direction.

Tear strength

[0069] Tear testing is done according to ASTM 1922. Machine direction is easier, as the thickness variation in critical test direction is better controlled. Thickness varies more in transverse direction and occasionally difficulties arise in taking the sample in a manner, which guarantees an even thickness for the critical testing area.

Water vapour transmission rate (WVTR)

[0070] Water vapour transmission rate was measured by using Permatran - W 100K water vapour permeation analysis system, commercially available from Modern Controls, Inc. (MOCON).

Basis weight

[0071] Basis weight can be determined in accordance with Federal Test Method No. 191A/5041. Sample size for the sample materials was 15.24x15.24 cm, and the resulting value is an average of at least three individual measurements.

Pinholes number

[0072] The presence of pinholes is determined by subjecting a film sample to water pressure corresponding to 650 mm water height.

[0073] The invention is further illustrated with the aid of the following examples.

Example 1 (Preparation of the catalyst)Complex preparation:

[0074] 87 kg of toluene was added into the reactor. Then 45.5 kg Bomag A in heptane was also added in the reactor. 161 kg 99.8 % 2-ethyl-1-hexanol was then introduced into the reactor at a flow rate of 24-40 kg/h. The molar ratio between BOMAG-A and 2-ethyl-1-hexanol was 1:1.83.

Solid catalyst component preparation:

[0075] 275 kg silica (ES747JR of Crossfield, having average particle size of 20 μm) activated at 600 °C in nitrogen was charged into a catalyst preparation reactor. Then, 411 kg 20 % EADC (2.0 mmol/g silica) diluted in 555 litres pentane was added into the reactor at ambient temperature during one hour. The temperature was then increased to 35 °C while stirring the treated silica for one hour. The silica was dried at 50 °C for 8.5 hours. Then 655 kg of the complex prepared as described above (2 mmol Mg/g silica) was added at 23 °C during ten minutes. 86 kg pentane was added into the reactor at 22 °C during ten minutes. The slurry was stirred for 8 hours at 50 °C. Finally, 52 kg TiCl_4 was added during 0.5 hours at 45 °C. The slurry was stirred at 40 °C for five hours. The catalyst was then dried by purging with nitrogen.

Example 2 (Preparation of the bimodal composition)

[0076] Into a 500 dm³ loop reactor, operated at 85 °C temperature and 60 bar pressure, was continuously introduced propane diluent, ethylene, hydrogen and 1-butene comonomer in such flow rates that ethylene content in the reaction mixture was 6.7 mol-%, the mole ratio of hydrogen to ethylene was 235 mol/kmol and the mole ratio of 1-butene to ethylene was 570 mol/kmol. At the same time into the reactor was continuously introduced a polymerisation catalyst prepared according to Example 1 and triethylaluminium cocatalyst in such quantities that ethylene polymer was produced at a rate of 25 kg/h. The molar ratio of aluminium of the cocatalyst to titanium of the catalyst was 20. The polymer had an MFR₂ of 300 g/10 min and density of 951 kg/m³.

[0077] The polymer was withdrawn from the loop reactor by using settling legs, and the polymer slurry was introduced into a flash tank operated at 3 bar pressure and 20 °C temperature.

[0078] From the flash tank the polymer was introduced into a fluidised bed gas phase reactor, which was operated at 80 °C temperature and 20 bar pressure. Into the gas phase reactor were additional ethylene, hydrogen and 1-butene introduced, as well as nitrogen flushes to keep the connections and piping open. Consequently, the concentration of ethylene in the reactor gas was 19 mol-%, the molar ratio of hydrogen to ethylene was 3 mol/kmol and the molar ratio of 1-butene to ethylene was 645 mol/kmol. The polymer was withdrawn from the reactor at a rate of 56 kg/h. After collecting the polymer, it was blended with additives and extruded into pellets in a counterrotating twin-screw extruder JSW CIM90P. The resulting polymer had an MFR₂ of 0.47 g/10 min and density of 922 kg/m³. The split between the polymer produced in the loop reactor and the polymer produced in the gas phase reactor was 45/55.

Example 3

[0079] The procedure of Example 2 was repeated, except that the conditions in the reactors were changed. The conditions and the resulting polymer data can be found in Table 1.

Example 4

[0080] The procedure of Example 2 was repeated, except that the conditions in the reactors were changed. The

conditions and the resulting polymer data can be found in Table 1.

Table 1

Example	2	3	4
Ethylene concentration in loop reactor, mol-%	6.7	6.7	6.7
Hydrogen to ethylene ratio in loop reactor, mol/kmol	235	265	305
1-butene to ethylene mole ratio in loop reactor, mol/kmol	570	514	0
Polymer production rate in loop reactor, kg/h	25	26	25
MFR ₂ of polymer produced in loop reactor, g/10 min	300	580	300
Density of polymer produced in loop reactor, kg/m ³	951	951	975
Ethylene concentration in gas phase reactor, mol%	19	7.8	8.2
Hydrogen to ethylene ratio in gas phase reactor, mol/kmol	3	7	8
1-butene to ethylene mole ratio in gas phase reactor, mol/kmol	645	460	480
Average particle size of the powder, mm	0.38	0.36	ND
MFR ₂ of the final polymer, g/10 min	0.47	0.21	ND
MFR ₂₁ of the final polymer, g/10 min	51	22	20
Density of the final polymer, kg/m ³	922	923	931
Split, loop/gpr	45/55	41/59	41/59
ND denotes that the respective property has not been determined			

Example 5

[0081] Polymer produced in Example 4 was compounded with SA233F (a high-impact copolymer of propylene with ethylene, produced and marketed by Borealis, having ethylene content of 14.5 % by weight and MFR₂, determined at 230 °C, of 0.8 g/10 min) and calcium carbonate. The final composition contained 35 % by weight of the bimodal polyethylene composition of Example 4, 10 % by weight of SA233F and 55 % by weight of CaCO₃. The thus obtained composition was then blown to a film and the resulting film was stretched in the machine direction 4.7 times its original length. The resulting film had a thickness of 30 µm, a basis weight of 34 g/m², tensile strength in the machine direction of 50 MPa, and in the transverse direction of 5 MPa. Tear strength in the machine and transverse directions were 1.2 and 40 N, respectively. The water vapour transmission rate was found to be 4990 g/m²/24 h. The film had no pinholes.

Example 6

[0082] The procedure of Example 5 was repeated, except that the polymer composition comprised of 40 % of polymer produced in Example 2 as the bimodal polyethylene composition and 60 % of CaCO₃. The composition was then blown to a film and the resulting film was stretched in the machine direction 6 times its original length. The resulting film had a thickness of 19 µm, a basis weight of 16 g/m², tensile strength in the machine direction of 59 MPa, and in the transverse direction of 4.1 MPa. Tear strength in the machine and transverse directions were 1.1 and 43 N, respectively. The water vapour transmission rate was found to be 6280 g/m²/24 h. The film had no pinholes.

Example 7

[0083] The procedure of Example 5 was repeated, except that the polymer composition comprised of 45 % of polymer produced in Example 3 as the bimodal polyethylene composition and 55 % of CaCO₃. The composition was then blown to a film and the resulting film was stretched in the machine direction 6 times its original length. The resulting film had a thickness of 25 µm, tensile strength in the machine direction of 67 MPa, and in the transverse direction of 4.1 MPa. Tear strength in the machine and transverse directions were 1.2 and 47 N, respectively.

Example 8

[0084] The procedure of Example 5 was repeated, except that the polymer composition comprised of 45 % of polymer

produced in Example 4 as the bimodal polyethylene composition and 55 % of CaCO_3 . The composition was then blown to a film and the resulting film was stretched in the machine direction 5 times its original length. The resulting film had a thickness of 28 μm , a basis weight of 26 g/m^2 , tensile strength in the machine direction of 86 MPa, and in the transverse direction of 6.0 MPa. Tear strength in the machine and transverse directions were 1.9 and 112 N, respectively. The water vapour transmission rate was found to be 1930 $\text{g/m}^2/24 \text{ h}$. The film had no pinholes.

Example 9

[0085] The procedure of Example 5 was repeated, except that the polymer composition comprised of 25 % of polymer produced in Example 4 as the bimodal polyethylene composition, 20 % of CB9270 (a bimodal linear low density polyethylene designed for extrusion coating, produced and marketed by Borealis, having a density of 927 kg/m^3 and MFR2 of about 10 g/10 min), and 55 % of CaCO_3 . The composition was then blown to a film and the resulting film was stretched in the machine direction 5 times its original length. The resulting film had a thickness of 21 μm , a basis weight of 23 g/m^2 , tensile strength in the machine direction of 72 MPa, and in the transverse direction of 6.2 MPa. Tear strength in the machine and transverse directions were 1.5 and 100 N, respectively. The water vapour transmission rate was found to be 1090 $\text{g/m}^2/24 \text{ h}$. The film had no pinholes.

Example 10

[0086] The procedure of Example 9 was repeated, except that film was stretched in the machine direction 5.5 times its original length. The resulting film had a thickness of 21 μm tensile strength in the machine direction of 85 MPa, and in the transverse direction of 5.5 MPa. Tear strength in the machine and transverse directions were 1.2 and 100 N, respectively.

Claims

1. A composition for making breathable films, the composition comprising:

(i) 20 - 50 %, based on the weight of the total composition, a bimodal polyethylene composition, further comprising

(ii) a first (low molecular weight) component, which is a homopolymer of ethylene or a copolymer of ethylene and one or more C_4 to C_{10} alpha-olefins, having a melt flow rate MFR_2 of 50 to 500 g/10 min , preferably of 100 to 400 g/10 min and a density of 940 to 975 kg/m^3 , preferably 945 to 975 kg/m^3 , the first component being present in the bimodal polyethylene composition in an amount of 37 to 48 % by weight, (iii) at least a second component, which is a copolymer of ethylene and one or more C_4 to C_{10} alpha-olefins, having a higher molecular weight, a lower melt index and a lower density than the said first component, the second component being present in the bimodal polyethylene composition in an amount of 52 to 63 % by weight, so that the said bimodal polyethylene composition has a melt flow rate MFR_2 in the range of 0.1 to 4.0 g/10 min , preferably 0.1 to 0.8 g/10 min , MFR_{21} in the range of 15 to 200 g/10 min , preferably 15 to 70 g/10 min and a density of 918 to 935 kg/m^3 ,

(iv) 40 - 70 %, based on the weight of the total composition, a particulate filler, and

(v) 0 - 30 %, based on the weight of the total composition, another olefin-based polymer

2. The composition according to Claim 1, wherein the other olefin based polymer is a propylene homo- or copolymer, preferably a copolymer of propylene and ethylene.

3. The composition according to Claim 2, wherein the composition comprises of 5 to 20 %, based on the weight of the total composition, of the said propylene polymer.

4. The composition according to Claim 1, wherein the content of the particulate filler is 55 to 70 %.

5. A composition according to any one of the preceding claims, wherein the particulate filler is calcium carbonate.

6. The use of the composition according to any one of the preceding claims, for making films.

7. Breathable polymer film, which film comprises a composition further comprising:

(i) 20 - 50 %, based on the weight of the total composition, a bimodal polyethylene composition, further comprising

- (ii) a first (low molecular weight) component, which is a homopolymer of ethylene or a copolymer of ethylene and one or more C_4 to C_{10} alpha-olefins, having a melt flow rate MFR_2 of 50 to 500 g/10 min, preferably of 100 to 400 g/10 min and a density of 940 to 975 kg/m³, preferably 945 to 975 kg/m³, the first component being present in the bimodal polyethylene composition in an amount of 37 to 48 % by weight,
- (iii) at least a second component, which is a copolymer of ethylene and one or more C_4 to C_{10} alpha-olefins, having a higher molecular weight, a lower melt index and a lower density than the said first component, the second component being present in the bimodal polyethylene composition in an amount of 52 to 63 % by weight, so that the said bimodal polyethylene composition has a melt flow rate MFR_2 in the range of 0.1 to 4.0 g/10 min, preferably 0.1 to 0.8 g/10 min, MFR_{21} in the range of 15 to 200 g/10 min, preferably 15 to 70 g/10 min and a density of 918 to 935 kg/m³,

(iv) 40 - 70 %, based on the weight of the total composition, a particulate filler, and

(v) 0 - 30 %, based on the weight of the total composition, another olefin-based polymer.

8. The film according to Claim 7, wherein the other olefin-based polymer is a propylene homo- or copolymer, preferably a copolymer of propylene and ethylene.

9. The film according to Claim 8, wherein the composition comprises of 5 to 20 %, based on the weight of the total composition, of the said propylene polymer.

10. The film according to Claim 7, wherein the content of the particulate filler is 55 to 70 %.

11. The film according to any one of Claims 7 to 10, wherein the particulate filler is calcium carbonate.

12. A film according to any one of Claims 7 to 11, wherein the film has a water vapour transmission rate of more than 3000 g/m²/24 h, preferably more than 4000 g/m²/24 h.

13. A film according to any one of Claims 7 to 12, wherein the film has a basis weight of less than 25 g/m².

14. A process for producing a breathable polymer film, comprising the steps of:

(A) providing into an extruder:

(i) 20 - 50 %, based on the weight of the total composition, a bimodal polyethylene composition, further comprising

- (ii) a first (low molecular weight) component, which is a homopolymer of ethylene or a copolymer of ethylene and one or more C_4 to C_{10} alpha-olefins, having a melt flow rate MFR_2 of 50 to 500 g/10 min, preferably of 100 to 400 g/10 min and a density of 940 to 975 kg/m³, preferably 945 to 975 kg/m³, the first component being present in the bimodal polyethylene composition in an amount of 37 to 48 % by weight,
- (iii) at least a second component, which is a copolymer of ethylene and one or more C_4 to C_{10} alpha-olefins, having a higher molecular weight, a lower melt index and a lower density than the said first component, the second component being present in the bimodal polyethylene composition in an amount of 52 to 63 % by weight, so that the said bimodal polyethylene composition has a melt flow rate MFR_2 in the range of 0.1 to 4.0 g/10 min, preferably 0.1 to 0.8 g/10 min, MFR_{21} in the range of 15 to 200 g/10 min, preferably 15 to 70 g/10 min and a density of 918 to 935 kg/m³,

(iv) 40 - 70 %, based on the weight of the total composition, a particulate filler, and

(v) 0 - 30 %, based on the weight of the total composition, another olefin-based polymer,

(B) extruding the composition to a film

(C) stretching the film to produce a breathable film.

15. The process according to Claim 14, wherein the other olefin-based polymer is a propylene homo- or copolymer, preferably a copolymer of propylene and ethylene.

16. The process according to Claim 15, wherein the composition comprises of 5 to 20 %, based on the weight of the total composition, of the said propylene polymer.

17. The process according to Claim 14, wherein the content of the particulate filler is 55 to 70 %.

18. The process according to any one of Claims 14 to 17, wherein the particulate filler is calcium carbonate.

19. The process according to any one of Claims 14 to 18, wherein the film is stretched with a stretching ratio of 3 to 10, preferably 4 to 7.

20. The process according to any one of Claims 14 to 19, wherein the bimodal polyethylene composition has been produced by:

(i) subjecting ethylene, hydrogen and optionally comonomers to a first polymerisation or copolymerisation reaction in the presence of the polymerisation catalyst in a first reaction zone to produce a first polymerisation product having a low molecular weight with a melt flow rate MFR_2 of 50 to 500 g/10 min, preferably of 100 to 400 g/10 min and a density of 940 to 975 kg/m³, preferably 945 to 975 kg/m³,

(ii) recovering the first polymerisation product from the first reaction zone,

(iii) feeding the first polymerisation product to a second reaction zone or reactor,

(iv) feeding additional ethylene, comonomers and optionally hydrogen to the second reaction zone,

(v) subjecting additional ethylene and additional comonomer(s) and optionally hydrogen to the second reaction zone in the presence of the said polymerisation catalyst and the first polymerisation product,

(vi) to produce a polymer composition comprising from 41 to 48 % by weight of the low molecular weight polymer produced in step (i), and from 59 to 52 % by weight of the high molecular weight component produced in step (v),

(vii) the composition having a melt flow rate in the range MFR_2 of 0.1 to 4.0 g/10 min, preferably 0.1 to 0.8 g/10 min and a density of 918 to 935 kg/m³, and

(viii) recovering the combined polymerisation product from the second reaction zone.



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EUROPEAN SEARCH REPORT

Application Number
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Place of search THE HAGUE		Date of completion of the search 20 November 2002	Examiner Schmidt, H
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**ANNEX TO THE EUROPEAN SEARCH REPORT
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